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The interplay between the statistical correlations of γ-ray emission probabilities and efficiency calibration

Ruy M. Castro[a,b,c], Vito R. Vanin[a,*], Otaviano Helene[a], Paulo R. Pascholati[a], Nora L. Maidana[a], Mauro S. Dias[d], Marina F. Koskinas[d]

[a] *Laboratorio do Acelerador Linear, Instituto de Fisica, Universidade de Sao Paulo, Rua do Matao, travessa R, 187, 05404-900 Sao Paulo, SP, Brazil*

[b] *Departamento de Matematica e Fisica, Universidade de Taubate, Rua Daniel Danelli s/n, 12060-440 Taubate, SP, Brazil*

[c] *Divisao de Sensoriamento Remoto, Instituto de Estudos Avançados, Centro Tecnico Aeroespacial, Rodovia dos Tamoios, km 5,5, 12228-840 S. Jose dos Campos, SP, Brazil*

[d] *Laboratorio de Metrologia Nuclear, Instituto de Pesquisas Energeticas e Nucleares, IPEN-CNEN/SP, Cidade Universitária, Rua do Matao, travessa R, 400, 05508-900 Sao Paulo, SP, Brazil*

Abstract

The procedure to determine the statistical correlations between γ-ray intensities is described here, as well as that required to take these correlations into account. The advantages of using branching-ratios and feeding fractions instead of γ-ray intensities in the γ-ray standards are discussed.

Keywords: X- and γ-ray spectroscopy; Radioactive source; Decay scheme

1. Introduction

The statistical correlations between γ-ray intensities are required in detector efficiency calibration when using multi-γ-ray radioactive sources for determining correctly standard deviations and performing statistical tests.

The γ-ray intensities of a multi-γ-ray radioactive source are correlated when they are determined simultaneously in an experiment where the detector efficiency is given by an analytical function fitted to experimental efficiency values. It requires many similar experimental studies to observe these correlations, consequently they are seldom evident. Nevertheless, the nuclide ^{152}Eu provides an example shown in Section 2.

The procedure to account for the statistical correlations in the efficiency calibration is described in Section 3 and the determination of the correlations between the γ-ray intensities due to the efficiency calibration is shown in Section 4. This approach provides a solution for the problem when there is no quest for the highest attainable precision and should be followed providing an appropriate framework for further data analysis. However, it is not the best solution because it leaves out the constraints imposed by the physical nature of the problem, the nuclear decay.

When developing γ-ray standards for detector efficiency calibration, the requirements of precision and consistency are higher, because all uncertainties will propagate in subsequent measurements. Overall consistency requires proper account of the decay scheme constraints, which leads to a reduction in the number of parameters and a corresponding increase in the number of degrees of freedom, resulting in better precision. The simplest approach is to fit the branching-ratios and feeding fractions to the observed peak areas in the γ-ray spectrum, as will be shown in Section 5. Besides, by obeying all decay-scheme constraints, branching-ratios and feeding fractions form the minimal data set that describes a decay scheme. The central problem of

*Corresponding author. Instituto de Fisica, Universidade de Sao Paulo, CP 66318, Sao Paulo, SP, 05315-970, Brazil. Tel.: +55-11-3091-6853; fax: +55-11-3091-6832.
E-mail address: vanin@if.usp.br (V.R. Vanin).

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calibrating the efficiency of a detector without any dependency on unknown statistical correlations is briefly presented in Section 6.

2. γ-Ray intensities correlations in ^{152}Eu

Sources of ^{152}Eu are often used in efficiency calibration because the emitted γ-rays cover a large energy range. In an effort to improve the precision of the decay data, many different laboratories determined the γ-ray intensities in the scope of an IAEA Coordinated Research Project (Bambynek et al., 1991). Fig. 1 presents scatter-plots of values for selected pairs of γ-ray transition intensities obtained by the different laboratories. From the plots it is possible to infer the existence of correlations and even estimate the correlation coefficient of the data set but, since the experiments were not planned to determine the correlations, it is impossible to determine exactly the correlation between the mean-values of the intensities, which requires a least-squares method (LSM) procedure.

3. Efficiency calibration taking into account the covariances

Detector efficiency calibration is almost invariably performed by LSM. In practice, a matrix formulation is required to deal with the covariances (Kendall et al., 1979; Eadie et al., 1971). In the following, we will summarize the formulas for the linear model, without loss of generality because the covariances enter into the calculations in the same way in the non-linear case. The general case of non-linear efficiency calibration is described elsewhere (IAEA, 2004; Venturini and Vanin, 1993).

3.1. The procedure

Consider a set of experimental values for the efficiencies determined from observed peak-areas A_j, each one corresponding to a γ-ray of energy E_j with intensity I_j photons per decay,

$$\varepsilon_j = \frac{A_j}{N \cdot I_j}, \tag{1}$$

where N is a normalization constant. It is assumed that A_j is the peak-area corrected for all secondary detection effects, including pile-up and sum (Debertin and Helmer, 1988; Mann et al., 1988; Knoll, 1989). The absolute efficiency is obtained with the substitution

$$N = \Omega \pm \sigma_\Omega,$$

where Ω is the number of decays and σ_Ω its standard deviation.

In most experiments, the three quantities in the right-hand side of Eq. (1) are statistically independent and the peak-areas are uncorrelated. Therefore, the covariance between two experimental values is given by

$$\frac{\mathrm{cov}(\varepsilon_j, \varepsilon_k)}{\varepsilon_j \varepsilon_k} = \left[\left(\frac{\sigma_{Aj}}{A_j}\right)^2 + \left(\frac{\sigma_{Ij}}{I_j}\right)^2 \right] \delta_{jk} + \left(\frac{\sigma_\Omega}{\Omega}\right)^2 + \frac{\mathrm{cov}(I_j, I_k)}{I_j I_k}, \tag{2}$$

where δ_{jk} is the Kronecker delta and the term involving Ω exists only for absolute calibrations.

A linear relationship between the efficiency parameters and the experimental data can be given by the matrix expression

$$\vec{y} = \mathbf{X} \cdot \vec{a}_0 + \vec{e}, \tag{3}$$

where $\vec{y}$ is the data vector, $\vec{a}_0^{\,t} = (a_{01}, a_{02}, \ldots, a_{0\mu})$ is the vector of parameters to be estimated, and the design matrix $\mathbf{X}$ is defined by $X_{iv} = \partial y_i / \partial a_v$. The quantity e_i is

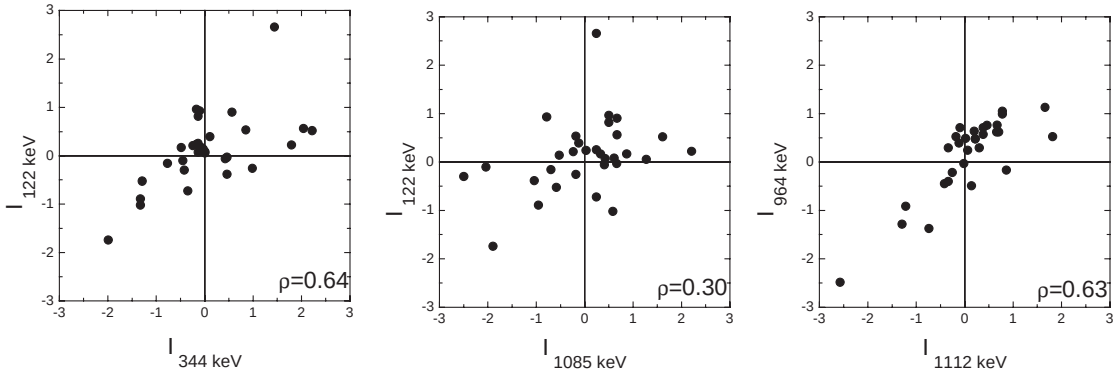

Fig. 1. Each plotted point corresponds to the emission probabilities of the pair of γ-ray transitions following ^{152}Eu decay whose energies label the axis, observed in each one of the 31 experiments performed in the scope of the IAEA CRP (Bambynek et al., 1991) The values plotted are the differences to the mean values, normalized by their respective standard deviations. The correlation coefficient of the data set, ρ is presented for each plot.

the unknown experimental error of y_i with $\langle e_i \rangle = 0$, $\langle e_i^2 \rangle = \sigma_i^2$, and $\langle e_i e_j \rangle = \mathrm{cov}(y_i, y_j)$. In order to make the discussion less abstract, let us assume that the efficiency is well represented by the function

$$\ln \varepsilon = \sum_{v=1}^{\mu} a_v \left(\ln \frac{E}{E_b} \right)^{v-1}, \tag{4}$$

where the reference energy E_b is conveniently chosen around the middle of the energy range; hence $y_i = \ln \varepsilon_i$, and the design matrix is defined through $X_{iv} = (\ln(E_i/E_b))^{v-1}$.

The LSM estimate of $\vec{a}_0, \hat{\vec{a}}$, is obtained by minimizing

$$Q(\vec{a}) = (\vec{y} - \mathbf{X} \cdot \vec{a})^{\mathbf{t}} \cdot \mathbf{V}^{-1} \cdot (\vec{y} - \mathbf{X} \cdot \vec{a}) \tag{5}$$

with respect to all a_v. In this case, allowing for the logarithmic transformation of the efficiency,

$$V_{jk} = V_{\ln \varepsilon_j \ln \varepsilon_k} = \frac{\mathrm{cov}(\varepsilon_j, \varepsilon_k)}{\varepsilon_j \varepsilon_k} \tag{6}$$

and, therefore, each element of $\mathbf{V}$ is given by the right-hand side of Eq. (2).

The solution of Eq. (5) is

$$\hat{\vec{a}} = (\mathbf{X^t} \cdot \mathbf{V^{-1}} \cdot \mathbf{X})^{-1} \cdot \mathbf{X^t} \cdot \mathbf{V^{-1}} \cdot \vec{y} \tag{7}$$

and the covariance matrix of the fitted parameters is given by

$$\mathbf{V}_a = (\mathbf{X^t} \cdot \mathbf{V^{-1}} \cdot \mathbf{X})^{-1}. \tag{8}$$

3.2. Correlation in interpolation

The efficiency for each γ-ray can be calculated from Formula (4) using the appropriate energy with the fitted parameters, giving a set of values ε_i whose covariance matrix can be calculated from the covariance matrix of the fitted parameters (Eq. (8)). The interpolated efficiency values for different energies are correlated, and cause the correlation between different γ-ray intensities as detailed in the next section. The propagation of the covariances from the efficiency calibration coefficients to the efficiencies at different energies can be condensed into a matrix formula,

$$\mathbf{V}_\varepsilon = \mathbf{D} \mathbf{V}_a \mathbf{D}^t, \tag{9}$$

where $\boldsymbol{V}_a$ is the covariance matrix of the fitted efficiency parameters (Eq. (8)) and $\mathbf{D}$ is the matrix of derivatives with respect to the parameters that, considering the efficiency model given in Eq. (4), results in

$$D_{jv} = \left. \frac{\partial \varepsilon(E_j)}{\partial a_v} \right|_{\hat{\vec{a}}} = \varepsilon(E_j) \left[\ln \left(\frac{E_j}{E_b} \right) \right]^{v-1}. \tag{10}$$

4. Determination of covariances between γ-ray intensities in spectroscopy measurements

This section outlines the calculation of the γ-ray intensity covariances in a usual γ-ray spectroscopy experiment. Even if the application example developed here is simplified, it contains all the required elements for the calculation of the covariance matrix in real cases. We made the discussion more concrete by choosing a simple decay scheme. However, the generalization is straightforward.

4.1. Data description

Consider the decay scheme of Fig. 2 and assume that the γ-rays γ_i were observed with a detector whose efficiency was calibrated using Eq. (4). For each γ_i, the observed peak-area is $C_i' \pm s_i'$ that, after correcting for sum, pile-up, and any other secondary detection effects, changes to $C_i \pm s_i$. Although the corrections introduce some statistical dependencies between the different values, the resulting covariances can normally be neglected. Therefore, the covariance matrix of the net peak areas can be well approximated by the diagonal matrix

$$\mathbf{V}_C = \begin{vmatrix} s_1^2 & & & & \\ & \ddots & & \mathbf{0} & \\ & & s_i^2 & & \\ & \mathbf{0} & & \ddots & \\ & & & & s_m^2 \end{vmatrix}, \tag{11}$$

where m is the number of γ-ray transitions. However, if there are unresolved doublets, the covariances between the corresponding peak areas must not be neglected and shall be placed in the appropriate rows and columns of $\mathbf{V}_C$.

The γ-ray intensities are calculated from Expression (1), where now the normalization constant is interpreted

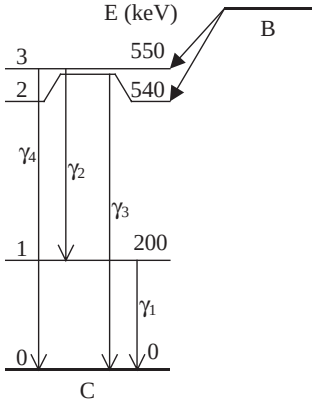

Fig. 2. Decay scheme used in the example of Sections 4 and 5.

differently and depends on the peak-areas, the efficiency, and possibly on a complimentary set of parameters τ (which includes the ground-state beta feeding fraction, internal conversion coefficients, etc.),

$$I_i = \frac{C_i}{N(C,\varepsilon,\tau)\varepsilon_i}. \tag{12}$$

For the sake of brevity, we will neglect the conversion coefficients and assume no beta feeding to the ground state.

4.2. Absolute γ-ray intensities

Considering that the sum of intensities of the transitions to the ground state is 1, the normalization factor for the decay scheme of Fig. 2 is determined as

$$N(C,\varepsilon) = \frac{C_1}{\varepsilon_1} + \frac{C_3}{\varepsilon_3} + \frac{C_4}{\varepsilon_4}. \tag{13}$$

Note that the determination of the emission intensities along with their covariances is a problem of change of variables, with m functions (given by Eq. (12)) of $2m$ random variables: m net peak-areas and m efficiencies. However, since the observed peak-areas and the interpolated efficiencies are statistically independent, the matrix of total variances of the intensities can be given by the sum of the variance matrices corresponding to these two primary data sets, avoiding the use of larger matrices to accommodate all $2m$ random variables at once,

$$\mathbf{V}_I = \mathbf{F}\mathbf{V}_C\mathbf{F}^t + \mathbf{G}\mathbf{V}_\varepsilon\mathbf{G}^t, \tag{14}$$

where $\mathbf{F}$ and $\mathbf{G}$ are defined through

$$F_{iv} = \frac{\partial I_i}{\partial C_v} \quad \text{and} \quad G_{iv} = \frac{\partial I_i}{\partial \varepsilon_v}, \tag{15}$$

respectively. Care must be taken when calculating these derivatives in order to capture the correct dependency on the random variables.

The result that reduces the experimental data, i.e., the set of values that summarizes and conveys all the statistical information obtained in the experiment in a way that no other data will be required for the calculation of any statistical quantity related to it, is the set of intensities I_i and their covariance matrix, $\mathbf{V}_I$.

4.3. Relative γ-ray intensities

Sometimes, however, only relative intensities are provided, either because the decay scheme is not sufficiently well known to find a suitable normalization factor or because they are much more precise than the absolute ones. In this case, the covariances between the normalization factor and the relative intensities must be provided to allow further statistical calculations, like normalizing the decay scheme or changing the reference line for intensities.

The normalization factor for relative intensities in this case is

$$N_r(C,\varepsilon) = \frac{C_r}{I_r\varepsilon_r}, \tag{16}$$

where r identifies the reference transition.

Since the reference value has no error, the dimension of the covariance matrix of the relative transition intensities is $m-1$, m being the number of γ-ray transitions. However, with this smaller matrix it will be impossible to retrieve the correct uncertainties in the relative photon fluxes $\phi_i = C_i/\varepsilon_i$, which are in number of m. The best way to complete the set of variables is to add the normalization factor N_r to the set of relative intensities and re-scale the normalization factor to the reference value I_r. This re-scaling is obtained by defining a constant Σ which equals the numeric value of N_r but which is not a random variable as N_r is; hence, the additional random variable to be considered when constituting the covariance matrix of the relative intensities is

$$I_{ref} = \frac{I_r N_r}{\Sigma}, \tag{17}$$

where we emphasize that Σ is not a random variable but has the value of the calculated normalization factor, much like I_r which is an arbitrary constant and not a random variable. Thus, I_{ref} is an exquisite variable with the value of the (arbitrary) reference intensity and the statistical behavior of the (random) normalization factor.

Note that, when restoring the original values for the relative photon fluxes, all the covariance terms must be taken into account. For example, when recalculating $\phi_1 = C_1/\varepsilon_1 = N_r I_{r1} \propto I_{ref} I_{r1}$, its relative variance is given by

$$\left(\frac{\sigma_{\phi_1}}{\phi_1}\right)^2 = \left(\frac{\sigma_{I_{ref}}}{I_{ref}}\right)^2 + \left(\frac{\sigma_{r1}}{I_{r1}}\right)^2 + 2\frac{\mathrm{cov}(I_{r1}, I_{ref})}{I_{r1} I_{ref}}.$$

5. Fitting the decay-scheme parameters

In the following, we will focus on the decay scheme shown in Fig. 2 to explain the method. All the formulas in this section, relating γ-ray intensities to the decay parameters with or without sum correction, can be obtained for any decay scheme in compact formulation (Andreev et al., 1972; Morel et al., 1985), which also takes into account the internal conversion coefficients (ICC), very important for the precise evaluation of a decay scheme. However, we will not include the ICCs in the formulas below because they just clutter the formulas, adding nothing new to the understanding of the problem.

5.1. Imposing the constraints

To take into account the decay-scheme constraints, Eq. (12) must be changed so that it expresses appropriately these constraints. Note that the intensities of the four γ-rays represented in Fig. 2 are not independent and can be written as functions of only two decay parameters, which we will call f_3, the beta feeding fraction to level 3, and κ_{3-1}, the branching-ratio from level 3 to 1:

$$\begin{aligned} I_1 &= f_3\kappa_{3-1}, \\ I_2 &= f_3\kappa_{3-1}, \\ I_3 &= f_2 = 1 - f_3, \\ I_4 &= f_3\kappa_{3-0} = f_3(1 - \kappa_{3-1}). \end{aligned} \tag{18}$$

Substituting the formulas above in Formula (12), we find a set of four relations between f_3, κ_{3-1}, and N, given by the four experimental peak-areas C_j, which allows the determination of these three parameters and their covariances by the LSM.

Note that the γ-ray intensities can be recalculated from the beta feeding fractions and branching-ratios by the same set of Eq. (18) used to impose the constraints; their variance matrix can be obtained by a procedure similar to that adopted for the efficiencies in Section 3.1.

5.2. Correcting for sum

In a real experiment, sum correction is most likely to be performed, either because a close geometry is used giving rise to an important correction, or because the precision required is high. In the example, the observed peak area for transition γ_1 is

$$A_1 = N\varepsilon_1\left[f_3\kappa_{3-1} - f_3\kappa_{3-1}\varepsilon_{total}(\gamma_2)\right],$$

where ε_{total} is the total efficiency. It can be pointed out that this expression, although more complicated than that given by Eq. (18), still depends on the same parameters, which can still be fit by the LSM. Moreover, if written in terms of γ-ray intensities, these expressions tend to be more complicated, because the branching-ratios are functions of the transition intensities.

5.3. Evaluating branching-ratios measured in decays from different parents

The intensities of γ-ray transitions are very sensitive to the specific nuclear reaction; therefore the branching-ratio measurement precision varies strongly with the parent nuclide. Averaging the branching-ratios from two different decays by taking into account the covariances must be accomplished by the LSM with the appropriated data vector and design matrix. Let us call $\vec{y}$ and $\vec{z}$ the data vectors containing the branching-ratios measured in the decays from nuclides Y and Z, respectively. Most likely, the dimensions of the data vectors will be different, because the levels fed by each of the two parents are different. Let us call $\vec{A}$ the vector of all branching-ratios observed in both decays. The model equations can be cast in a format similar to Eq. (3),

$$\begin{pmatrix} \vec{y} \\ \vec{z} \end{pmatrix} = \mathbf{X}\vec{\kappa}_0 + \vec{\varepsilon}, \tag{19}$$

where just one element of each row of $\mathbf{X}$ is 1, that in the column corresponding to the index of the branching-ratio in $\vec{\kappa}_0$, all the other elements in the row being null. The variance matrix of the data vector is also constituted of blocks,

$$\mathbf{V}_{yz} = \begin{pmatrix} \mathbf{V}_y & \mathbf{0} \\ \mathbf{0} & \mathbf{V}_z \end{pmatrix}. \tag{20}$$

The averages and the covariance matrix are calculated by formulas (7) and (8), using the $\mathbf{X}$ and $\mathbf{V}_{yz}$ given in Eqs. (19) and (20).

6. Discussion

All the elements for the correct determination and use of the statistical correlations between γ-ray intensities determined in spectroscopy measurements are well known and well studied. However, their integration is cumbersome and requires proper bookkeeping. Unfortunately, due to the nature of the problem, there is no simpler solution than that presented here.

Since the γ-ray intensity covariances are required to determine the covariances between the efficiencies, which in turn are required to determine the γ-ray intensity covariances, there is a circular dependency in these calculations that must be broken. One possibility is to calibrate the detector efficiency using activity-calibrated monochromatic sources, which is a very difficult task because it requires many activity-calibrated standard sources simultaneously. Other possibility consists in calculating the efficiency by simulation guided by a few measurements of selected activity-calibrated standard sources (Hardy et al., 2002; Helmer et al., 2003).

When developing γ-ray standards for detector calibration, it is possible, with some additional effort, to determine the branching-ratios and feeding fractions instead of γ-ray intensities, which has many advantages: they form the minimal data set that specifies completely a decay scheme; they embody the constraints arising from the nature of the physical problem; their determination requires the complete specification of the decay properties, including conversion coefficients, leaving no room for ambiguities in the application of the standard; they are the required quantities to correct for summing, necessary in almost any real-life efficiency calibration.

Finally, branching-ratios can be easily averaged when observed in the decay of different isobars and isomers, improving the accuracy of the decay data.

7. Conclusion

Section 4 presented the procedure to determine the covariances between γ-ray intensities in a usual γ-ray spectroscopy measurement. Unless the covariances between the standard emission probabilities are well determined, this procedure should be followed assuming that these covariances are negligible. This is a good approximation because most of the statistical dependency between the intensities arises from the normalization and the common efficiency calibration curve, appropriately accounted for by Eq. (14). The procedure outlined in Section 5 is proposed to develop better decay data standards and should not be applied in general.

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