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INSTITUTO DE FÍSICA  
CAIXA POSTAL 20516  
01498-970 SÃO PAULO - SP  
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GAMMA-RAY CALIBRATION ENERGIES: A REVIEW  
OF THE  $^{192}\text{Ir}$  DATA

O. Helene and Vito R. Vanin

Instituto de Física, Universidade de São Paulo

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Gamma-ray calibration energies: a review of the  $^{192}\text{Ir}$  data

O. Helene and Vito R. Vanin.

Instituto de Fisica da Universidade de Sao Paulo, C.P. 20516,  
01498 Sao Paulo, BRAZIL.

In order to perform calibrations by the least squares method, the complete covariance matrix for the gamma-ray energy standards is needed. Some errors due to excluding the covariances in the statistical analysis are pointed out. The gamma-ray energies of  $^{192}\text{Ir}$  and  $^{198}\text{Au}$  measured with curved crystals spectrometers were re-analyzed and the covariance between results found. The gamma-ray energies were updated to include the latest values of the fundamental constants. The covariance matrix between gamma-ray energy data and the fundamental constants is deduced. The covariance matrices are indispensable to update the energy values if new measurements are performed or if the fundamental constants are reevaluated.

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## I. Introduction

Covariances between experimental results are as significant as variances in the evaluation of experimental uncertainties. Furthermore, covariances are indispensable for updating experimental results. If two quantities are correlated and one of them has been re-evaluated, the other must be updated, which can be done only if their covariance is known.

The covariance, or correlation, between two results originates in the experimental procedure. If an error source affects both results then they are correlated. When the covariance between two results is positive (negative) and one of them is overestimated then the other one is probably overestimated (underestimated).

When dealing with correlated data, a full account of covariances calls for a matrix treatment of the data (see Appendix), with a Variance Matrix where the diagonal elements represent the variances, and the off diagonal elements represent the covariances.

Among the effects due to the omission of covariances are unreliable confidence intervals and chi-square values.

Energies for  $^{192}\text{Ir}$  and  $^{198}\text{Au}$  have been updated by the recent reevaluation of fundamental constants [1]. It will be supposed that the covariance of functions of random variables can be approximated by the usual linear formula, since uncertainties are small when compared with mean values. When a probability is assigned to a confidence interval or to the result of a statistical test, the density functions for the data will be assumed to be Gaussian.

## II. Basic concepts.

This section shows some basic results and some examples about the use of covariances.

(i) Consider two quantities  $y_1$  and  $y_2$  with variances  $\sigma_1^2$  and  $\sigma_2^2$ , respectively, and covariance  $\text{cov}(y_1, y_2) = \rho\sigma_1\sigma_2$ , where  $\rho$  is the

correlation coefficient. The variance of  $a = y_1 + y_2$  is

$$\sigma_a^2 = \sigma_1^2 + \sigma_2^2 + 2\rho\sigma_1\sigma_2. \quad (1)$$

The omission of the covariance in this equation leads to an erroneous value of the standard deviation of  $a$ .

(ii) Assume now that  $y_1$  and  $y_2$  are the results of two observations of the same quantity with true value  $a_0$ , which is unknown. The least squares estimate of the measured quantity is (see item(a) of Appendix)

$$\bar{a} = \frac{y_1(\sigma_2^2 - \rho\sigma_1\sigma_2) + y_2(\sigma_1^2 - \rho\sigma_1\sigma_2)}{\sigma_1^2 + \sigma_2^2 - 2\rho\sigma_1\sigma_2} \quad (2)$$

with variance

$$\sigma_a^2 = \frac{\sigma_1^2\sigma_2^2(1 - \rho^2)}{\sigma_1^2 + \sigma_2^2 - 2\rho\sigma_1\sigma_2} \quad (3)$$

The omission of the covariance leads to wrong estimates of both the quantity and its variance. When  $\sigma_2 < \rho\sigma_1$  or  $\sigma_1 < \rho\sigma_2$ , equation (2) gives a negative weight for  $y_1$  or  $y_2$ . This is an inevitable consequence of the correct use of covariances and should not be considered an anomaly[2,3].

(iii) If two quantities are correlated and only one of them is measured in a further experiment, the estimate of the other must also be reviewed. This can be accomplished only if the whole covariance matrix is known.

Suppose that two quantities  $a_1$  and  $a_2$  were measured, being their estimates (and standard deviations)  $y_1(\sigma_1)$  and  $y_2(\sigma_2)$ , respectively, and covariance  $\text{cov}(y_1, y_2) = \rho\sigma_1\sigma_2$ . Suppose that another measurement of  $a_1$  gives the result  $y_1'(\sigma_1')$  which is correlated neither with  $y_1$  nor with  $y_2$ . This second measurement affects both estimates and we will show that the effect on the estimate of  $a_2$  is comparable to that on  $a_1$ . The least-squares estimates of  $a_1$  and  $a_2$  are

$$\bar{a}_1 = \frac{y_1/\sigma_1^2 + y_1'/\sigma_1'^2}{1/\sigma_1^2 + 1/\sigma_1'^2} \quad \text{and} \quad (4)$$

$$\bar{a}_2 = y_2 + \frac{(y_1' - y_1)\rho\sigma_1\sigma_2}{\sigma_1^2 + \sigma_1'^2} \quad (5)$$

with covariance matrix

$$V = \frac{1}{\sigma_1^2 + \sigma_1'^2} \begin{pmatrix} \sigma_1^2\sigma_1'^2 & \rho\sigma_1\sigma_2\sigma_1'^2 \\ \rho\sigma_1\sigma_2\sigma_1'^2 & \sigma_2^2[\sigma_1^2(1-\rho^2) + \sigma_1'^2] \end{pmatrix}, \quad (6)$$

where  $V_{11}$  and  $V_{22}$  are the variances of  $\bar{a}_1$  and  $\bar{a}_2$ , respectively, and the off-diagonal element is the covariance.

Equation (4) is the usual weighted average of two uncorrelated observations  $y_1$  and  $y_1'$ . If  $\rho=0$ , equation (5) shows that the estimate of  $a_2$  depends only on the result of the first measurement, which is almost obvious. The non intuitive result brought by equation (5) is that, if  $y_1$  and  $y_2$  are correlated then the estimate of  $a_2$  depends on the new measurement of  $a_1$ . In order to see the importance of this modification, let us assume that  $\sigma_1 = \sigma_2 = \sigma_1'$  and  $\rho=0.5$ . In this case we have

$$\bar{a}_1 = y_1 + \delta/2 \quad \text{and} \quad (7)$$

$$\bar{a}_2 = y_2 + \delta/4 \quad (8)$$

where  $\delta = y_1' - y_1$ . That is, both the estimates of  $a_1$  and  $a_2$  are changed by terms of the same magnitude.

(iv) Consider the fitting of a straight line  $y = a_1 + a_2x$  to experimental data  $\{(x_i, y_i), i=1, 2, \dots, n\}$ , where the variances of  $y_i$  are

all equal to  $\sigma^2$  and  $\text{cov}(y_i, y_j) = \rho\sigma^2$  for every  $i \neq j$ , and the independent variable  $x$  is measured without error. The adjusted value  $y_c$  of  $y$  at the mean value  $x_c$  of the  $x_i$  has variance

$$\sigma_c^2 = \sigma^2 \left( \frac{\rho(n-1)}{n} + \frac{1}{n} \right) \xrightarrow{n \rightarrow \infty} \rho\sigma^2 \quad (9)$$

This equation shows that the improved precision in the interpolation is limited by the covariance between the data. If the off-diagonal terms of the covariance matrix were omitted the (wrong) variance of  $y_c = y(x_c)$  would be written as

$$\sigma_c^2 = \frac{\sigma^2}{n} \xrightarrow{n \rightarrow \infty} 0 \quad (10)$$

The omission of the covariances leads to an unrealistic high precision of the measurement.

v) There are some cases where an earlier result is substituted by a new one, changing the adopted values of every correlated data. Consider, for example, two results  $y_1(\sigma_1)$  and  $y_2(\sigma_2)$  with covariance  $\rho\sigma_1\sigma_2$ . Suppose that  $y_2(\sigma_2)$  is substituted by  $y'_2(\sigma'_2)$  not correlated neither with  $y_1$  nor with  $y_2$ . Then  $y_1$  must be substituted by (see Appendix)

$$y'_1 = y_1 + \frac{\rho\sigma_1}{\sigma_2}(y'_2 - y_2) \quad (11)$$

The variance of  $y_1'$  becomes

$$\sigma_1'^2 = \sigma_1^2(1-\rho^2) + \rho^2 \left( \frac{\sigma_1}{\sigma_2} \right)^2 \sigma_2'^2 \quad (12)$$

and

$$\text{cov}(y'_1, y'_2) = \rho \frac{\sigma_1}{\sigma_2} \sigma_2'^2 \quad (13)$$

If  $\rho=0$  the new value  $y'_2$  does not change the older result:  $y'_1 = y_1$  and

$\sigma_1' = \sigma_1$ .

Such case occurs in gamma-ray spectroscopy since every gamma-ray energy is correlated with fundamental constants.

vi) In gamma-ray spectroscopy with curved crystals spectrometers it is usual to add uncertainties due to calibration, scale, lattice spacing, and divergence, to statistical uncertainties, without specification of the covariances. The present procedure aims to obtain both self-consistent results and consistency between different experiments. This concerns cases i) to v) above and will be developed in more detail in the next section

### III. Revisiting $^{192}\text{Ir}$ and $^{198}\text{Au}$ gamma-ray energies

Lorenz [4] presents the estimates and standard deviations of the energies of eleven gamma-ray transitions following  $^{192}\text{Ir}$  decay. Among them there are four cascade/cross-over relations (table 1). However, the standard deviations of the differences between the cross-over energy and the sum of the cascade energies are between 70 and 10000 times larger than the energy differences. This shows that the standard deviations are overestimated or that the covariances are not negligible. As we will see below, both causes exist.

The values of the gamma-ray energies of  $^{192}\text{Ir}$  used in this paper are taken from the experiments of Kessler et. al. [5] and Borchert et. al. [6]. Other measurements will not be considered here because they do not give sufficient details on the estimation of the uncertainties.

Data from Kessler et. al. [5] are reproduced in table (2). The standard deviations are those identified as "total uncertainties" in ref. [5]. The covariances for  $E_k \neq E'_k$  are given by

$$\text{cov}_0(E_k, E'_k) = \sigma_\alpha^2 E_k E'_k \quad (14)$$

where  $\sigma_\alpha$  corresponds to the quadratic addition of the relative standard deviations due to lattice spacing ( $0.2 \times 10^{-6} = 0.2$  ppm),

calibration (0.1 ppm) and vertical divergence (0.2 ppm) quoted by Kessler et. al. [5], giving

$$\sigma_{\alpha} = 3.10^{-7}. \quad (15)$$

Table 3 presents the data from Borchert et. al. [6]. Uncertainties in table 3 correspond to  $dE$  in ref. [6]. Although the uncertainties included in  $dE$  were not explicitly given in ref. [6], the experimental procedure adopted [7] suggests that  $\rho_B \approx 0.5$ . Three cascade/cross-over relations among the gamma-rays from ref. [6] give reduced chi-square 1 only if  $\rho_B = 0.58$ , which is used in this paper. However, any value in the range 0.4 through 0.7 would give practically the same final results.

#### IV. Updating energy data.

Kessler et. al. determined energies using the energy-wavelength conversion coefficient  $f_0 = hc/e = 1.2398520 \cdot 10^{-6}$  eV·m, assumed exact [5]. Thus, those data must be updated using the more recent values of the fundamental constants [1],  $f = 1.23984244(37) \cdot 10^{-6}$  eV·m, and propagating its uncertainty. Energy data from ref. [5], then, must be multiplied by

$$r = f/f_0. \quad (16)$$

The variance of data from ref. [5] is

$$\sigma^2(E_K) = \sigma_0^2(E_K) + \left( \frac{\sigma_f}{f} \right)^2 E_K^2, \quad (17)$$

where  $E_K$  is the transition energy and  $\sigma_0(E_K)$  is the original standard deviation from ref. [5], reproduced in table (2). For  $E_K \neq E'_K$ , the covariances between the updated data are

$$\text{cov}(E'_K, E_K) = \text{cov}_0(E'_K, E_K) + \left( \frac{\sigma_f}{f} \right)^2 E'_K E_K, \quad (18)$$

with  $\text{cov}_0(E'_K, E_K)$  calculated by formula (14) above.

Energies from ref. [6], reproduced in table 3, were measured using the 412keV gamma transition of  $^{198}\text{Au}$  as reference, assumed as 411.794 keV exactly. In order to update results from ref. [6] with respect to the value  $f$  and make them compatible with data from ref [5] they must be multiplied by

$$r = \frac{411.80441}{411.794} \frac{f}{f_0}, \quad (19)$$

where the value 411.80441 keV is the  $^{198}\text{Au}$  transition energy given by Kessler et. al.. The variances of the gamma-ray energies then become

$$\sigma^2(E_B) = \sigma_0^2(E_B) + \left\{ \left( \frac{\sigma_0(E_A)}{E_A} \right)^2 + \left( \frac{\sigma_f}{f} \right)^2 \right\} E_B^2, \quad (20)$$

where  $E_A$  is the energy of the 412 keV transition of  $^{198}\text{Au}$  and  $\sigma_0(E_A)$  its standard deviation shown in table (2).  $\sigma_0(E_B)$  are the standard deviations of gamma-ray energies, given in table (3). Covariances between the updated energies from Borchert et. al. are given by

$$\text{cov}(E'_B, E_B) = \rho_B \sigma_0(E'_B) \sigma_0(E_B) + \left\{ \left( \frac{\sigma_0(E_A)}{E_A} \right)^2 + \left( \frac{\sigma_f}{f} \right)^2 \right\} E'_B E_B. \quad (21)$$

The covariances between the original data from refs. [5] and [6], null before the updating, became non-zero after the updating due to the common dependence on  $f$  and the compatibility correction expressed by formula (19) above. They are given by

$$\text{cov}(E'_B, E_K) = \left( \frac{\sigma_f}{f} \right)^2 E'_B E_K + \frac{E'_B}{E_A} \text{cov}_0(E'_K, E_A), \quad (22)$$

where  $E'_K$  and  $E_B$  represent data from Kessler et. al. and Borchert et. al. and  $E_A$  is the energy of the 412 keV transition from  $^{198}\text{Au}$  measured by ref. [5]. When  $E'_K = E_A$ ,  $\text{cov}_0(E'_K, E_A)$  must be interpreted as  $\sigma_0^2(E_A)$  for expression (22) to remain valid.

The covariance between every transition from  $^{192}\text{Ir}$  and the factor  $f$  is

$$\text{cov}(E, f) = E \cdot f \cdot \left( \frac{\sigma_f}{f} \right)^2 \quad (23)$$

Although these covariances are not needed for this adjustment of the gamma-ray transition energies, they are necessary for a further updating of data due, for instance, to new measurements of the fundamental constants.

Data from refs. [5] and [6] must be multiplied by the ratio of the Si spacing used by Kessler et. al. [5] to that determined by the 1986 evaluation of physical constants [1],

$$\alpha = 1.00000138 \quad (24)$$

Since the statistical uncertainty of the lattice spacing of Si was not changed in the 1986 review and it was already included in the standard deviations of tables (2) and (3), the estimated variances do not have to be changed. The covariances between  $^{192}\text{Ir}$  gamma-rays and the lattice parameter of Si are given by

$$\text{cov}(E, a_0) = E \cdot a_0 \cdot \left( \frac{\sigma_{a_0}}{a_0} \right)^2 \quad (25)$$

where  $\sigma_{a_0} / a_0 = 0.2 \cdot 10^{-6}$  [1]. As eq. (23) above, eq. (25) may be needed in further updating of gamma-ray energies.

Figure (1) shows, in a schematic form, the variance matrix of the analyzed data.

There are 14 parameters to be fitted: 13 gamma-ray transition energies from  $^{192}\text{Ir}$  and the energy of the 412 keV transition of  $^{198}\text{Au}$ . Table (4) shows the energy data and the design matrix used in the fit by the least-squares method (see Appendix).

Table (5) shows the obtained results and table (6) shows the covariances and correlation coefficients of the fitted gamma-ray energies of  $^{192}\text{Ir}$  and the 412 keV of  $^{198}\text{Au}$ . The covariances between gamma-ray energies and  $f$  and  $a_0$  are given by equations (23) and (25) above. The chi-square value of the fit is  $\chi^2=12.8$  with 9 degrees of freedom, with a probability of being exceeded equals to about 20% .

## V. Discussion.

It is interesting to study here the consequences of the fit of correlated data in a real case. The results are not exactly given by the formulas from examples i to iv of section II since here we are selecting only two or three data from a larger correlated set. Anyway, they will prove their usefulness in displaying the main characteristics.

The standard deviations of the 136 keV, 468 keV and 604 keV gamma-rays shown in table (5) are respectively 0.48 eV, 0.28 eV and 0.49 eV. If covariances are neglected, the standard deviation of the cascade/cross-over relation 136keV+468keV-604keV would be 0.74 eV. Considering the covariance that standard deviation is 0.66 eV, shown in table (7). The overestimation of the standard deviation by neglecting the covariance was discussed in item ii, Section II.

The example of item iii can be seen in the case of some gamma-rays measured just in one of the two experiments considered: 136 , 416 , 201 and the 412 keV gamma-ray energies. In the first three cases the standard deviations are significantly reduced in the fit. In all cases the fitted values, Table 5, are different from the input values, Table 4, since the measurement of a quantity changes all correlated quantities.

In order to verify the discussion of item iv of section II we can observe that the standard deviations of the 412 keV and of the 205 keV are not reduced in the fit. This is due to the fact that the relative standard deviation of  $f$ ,  $0.3 \cdot 10^{-6}$ , imposes a lower limit in the uncertainties.

Table (7) shows the energy differences of three cascade/cross-over relations among the fitted data. These differences are of magnitude comparable with their standard deviations. The footnote b of table (7) shows the standard deviations calculated without the covariance terms, giving an example of the discussion of item i. Although the cascade/cross-over energies differences in table (7) are larger than the same differences in table (1), those results are statistically acceptable while the ones of table (1) are

unacceptably low.

In the following, we will discuss some consequences of another choice of the parameter  $\rho_B$ . For every  $\rho_B \leq 0.7$ , the probability of exceeding the  $\chi^2$  obtained in the final fit is higher than 5% and only for  $\rho_B > 0.8$  it is unacceptably low (smaller than 1%). The fitted energies are weakly dependent on  $\rho_B$ , except for the 316 keV gamma ray, which varies slowly from 316.50600 keV for  $\rho_B = 0.1$  to 316.50622 keV for  $\rho_B = 0.9$ . The dependence of the standard deviations of the fitted energies is noticeable only for the energies which were measured only by Borchert et al. [6]. For instance, the standard deviation of the 136 keV gamma-ray varies smoothly from 0.69 eV for  $\rho_B = 0.1$  to 0.25 eV for  $\rho_B = 0.9$ . Since the differences in the results for  $\rho_B$  in the range 0.4 to 0.7 are small, the uncertainty of  $\rho_B$  pointed out in section III is of no consequence.

## VI. Conclusion.

The fitted energy of the gamma-ray of  $^{198}\text{Au}$  is 411801.75 (19) eV. This value agrees with that proposed by Wapstra [8], 411801.85(15) eV.

Covariance matrices allow updating whenever new data become available.

With semi-conductor detectors, it is possible to measure gamma-ray energies within less than 1 eV precision if an ensemble of precise energy standards with their covariance matrices are known. Table 5 presents such a set of energy data for  $^{192}\text{Ir}$  and  $^{198}\text{Au}$ .

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## APPENDIX

### Least-squares method: use of the design matrix and the covariance matrix

This appendix shows the principal equations of the least-squares method applied to a linear model and used in this paper. References [9] and [10] also illustrate some cases where the same physical quantity was measured in different experiments and then adjusted to a common value. Proofs and examples can be found in [11] and [12].

Suppose a function  $f(\mathbf{x}; \mathbf{a})$  where  $\mathbf{x}$  stands for a  $m$  components independent variables vector and  $\mathbf{a}$  for a  $n$  parameters vector to be fitted. Suppose that the function  $f$  has been measured for  $N$  values of the independent variables obtaining experimental data  $y_i = f(\mathbf{x}_i; \mathbf{a}_0) + \varepsilon_i$ , where  $\mathbf{a}_0$  is the true, unknown value of the parameters vector, and  $\varepsilon_i$  is the experimental error of the measurement of  $y_i$ . The experimental data and the parameters are related by

$$\mathbf{Y} = \mathbf{X} \cdot \mathbf{A}_0 + \vec{\varepsilon} \quad , \quad (A1)$$

where  $\mathbf{Y}$  is the (column) vector whose elements are the experimental data  $y_i$ ,  $\mathbf{A}_0$  is the vector whose elements are the true values of the parameters,  $\vec{\varepsilon}$  is the unknown vector of errors, and  $\mathbf{X}$  is the design matrix.  $\mathbf{X}$  is independent of  $\mathbf{A}_0$  and contains all the dependence on  $\mathbf{x}$ .

Two assumptions are needed in order to calculate the least-squares estimate: the measurements are unbiased,

$$\langle \varepsilon_i \rangle = 0 \quad ,$$

and the variance matrix, given by

$$V_{ij} = \langle \varepsilon_i \varepsilon_j \rangle \quad , \quad (A2)$$

is known, where  $\langle \rangle$  stands for the expectation value. The least-squares estimate of  $\mathbf{A}$  is given by

$$\tilde{\mathbf{A}} = (\mathbf{X}^t \mathbf{V}^{-1} \mathbf{X})^{-1} \mathbf{X}^t \mathbf{V}^{-1} \mathbf{Y} \quad , \quad (A3)$$

with covariance matrix given by

$$W_A = (X^t W^{-1} X)^{-1} \quad (A4)$$

The statistics calculated by

$$\chi^2 = (Y - X\bar{A})^t W^{-1} (Y - X\bar{A}) \quad (A5)$$

has a chi-square probability density function (pdf) corresponding to N-n degrees of freedom when the pdf of the errors is a N dimensional gaussian.

It can be shown that the estimate (A3) is unbiased, is the linear estimate of minimum variance, and that (A4) gives the exact variance matrix independent of the error pdf, as long as W is known exactly. Also, the expectation of  $\chi^2$  given by (A5) equals N-n, independent of the error pdf, but its pdf is not the usual chi-square unless the errors are gaussian distributed. Finally, the pdf of the estimates  $\bar{A}_j$  are gaussian only when the errors are gaussian or the number of data points, N, is enough to fulfill the Central Limit theorem conditions in the statistics given by (A3).

This method is applied in two particular cases of interest (a, b below). Item c discusses how to update data when a result is substituted by a new one.

a) Two correlated measurements of the same physical quantity.

Suppose that a physical quantity has been measured twice, with the results  $Y_1$  and  $Y_2$ , and covariance matrix W. In this case  $f(x_1) = f(x_2) = a$ , giving

$$X = \begin{pmatrix} 1 \\ 1 \end{pmatrix} \quad (A6)$$

From equation (A3) we obtain the value  $\bar{a}$  given by equation (2) of the text. Equation (A4) gives the result of eq. (3) of the text.

b) Two measurements of the same physical quantity, one of them covariant with another quantity.

Suppose two quantities,  $a_1$  and  $a_2$ , whose experimental values are  $y_1$  and  $y_2$ , respectively, being W the covariance matrix. Suppose that a new measurement of  $a_1$  gives the result  $y_1'$  with variance  $\sigma_1'^2$ , statistically independent of  $y_1$  and  $y_2$ . The three experimental results may be written as the vector

$$Y = (y_1, y_2, y_1') \quad (A7)$$

The variance matrix of Y is

$$W_Y = \left( \begin{array}{c|c} W & 0 \\ \hline 0 & \sigma_1'^2 \end{array} \right) = \begin{pmatrix} \sigma_1^2 & \rho\sigma_1\sigma_2 & 0 \\ \rho\sigma_1\sigma_2 & \sigma_2^2 & 0 \\ 0 & 0 & \sigma_1'^2 \end{pmatrix} \quad (A8)$$

In this case we have  $f(x_1) = a_1$ ,  $f(x_2) = a_2$  and  $f(x_3) = a_1$ , giving a design matrix X

$$X = \begin{pmatrix} 1 & 0 \\ 0 & 1 \\ 1 & 0 \end{pmatrix} \quad (A9)$$

Substituting equations (A7) - (A9) in equation (A3) we obtain the results indicated in formulas (4) and (5) of the text. Equation (A4) gives the covariance matrix of formula (6) of the text.

c) Substitution of standards

Consider three estimates  $Y_1, Y_2, Y_3$  whose variance matrix is V. Suppose that a new experiment measures one of those quantities and the old result is substituted by the new one that is not correlated with  $Y_1, Y_2$  and  $Y_3$ . Assume that  $y_3$  is substituted by  $y_3'$  with variance  $\sigma_3'$ . In this case  $y_1$  and  $y_2$  must be updated by



$$y'_j = y_j + \frac{\text{cov}(y_j, y_3)}{\sigma_3^2} (y'_3 - y_3), \quad j=1,2, \quad (\text{A10})$$

where  $\text{cov}(y_j, y_3) = V_{j3}$  is the covariance between  $y_j$  and  $y_3$  and  $\sigma_3^2 = V_{33}$  is the variance of  $y_3$ . Eq (A10) corresponds to new estimates of the first two quantities obtained from the conditional probability density function of  $y_1, y_2$  and  $y_3$  for  $y_3 = y'_3$ .

The covariance matrix of  $y'_1, y'_2$  and  $y'_3$  is obtained from the following procedure:

i) the covariance matrix  $V_R$  of  $y'_1, y'_2$  is obtained by removing the 3<sup>rd</sup> row and column of  $V^{-1}$  and inverting the resultant 2x2 sub-matrix;

ii) the variance matrix  $W'$  of  $y'_1, y'_2, y'_3$  is given by

$$V' = \begin{pmatrix} V_R & 0 \\ 0 & 0 \end{pmatrix} + \Delta V, \quad (\text{A11})$$

where

$$\Delta V = \begin{pmatrix} \left( \rho_{13} \frac{\sigma_1}{\sigma_3} \right)^2 & \frac{\rho_{12} \rho_{23} \sigma_1 \sigma_2}{\sigma_3^2} & \rho_{13} \frac{\sigma_1}{\sigma_3} \\ \frac{\rho_{12} \rho_{23} \sigma_1 \sigma_2}{\sigma_3^2} & \left( \rho_{23} \frac{\sigma_2}{\sigma_3} \right)^2 & \rho_{23} \frac{\sigma_2}{\sigma_3} \\ \rho_{13} \frac{\sigma_1}{\sigma_3} & \rho_{23} \frac{\sigma_2}{\sigma_3} & 1 \end{pmatrix} \sigma_3'^2. \quad (\text{A12})$$

This procedure is an uncertainty propagation formula and can be generalized to any number of data.

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Table 1 - Four cascade/cross-over relations between gamma-rays from  $^{192}\text{Ir}$  decay.  $E_1$  and  $E_2$  are the energies of two consecutive gamma-rays and  $E_{co}$  the energy of the corresponding cross-over.  $\delta E$  stands for the cascade/cross-over energy difference,  $\delta E = E'_1 + E'_2 - E'_{co}$ , where the primes are energies corrected for recoil.  $\sigma_E$  is the standard deviation of  $\delta E$ . Data from [4].

| $E_1$ (keV) | $E_2$ (keV) | $E_{co}$ (keV) | $\delta E$ (eV) | $\sigma_E$ (eV) <sup>a</sup> |
|-------------|-------------|----------------|-----------------|------------------------------|
| 136.3434    | 468.0715    | 604.41455      | -0.006          | 2.1                          |
| 295.95821   | 308.45685   | 604.41455      | 0.000           | 2.0                          |
| 295.95821   | 316.50800   | 612.46569      | -0.004          | 2.0                          |
| 295.95821   | 588.5851    | 884.5423       | 0.036           | 2.7                          |

a) Square root of the sum of the variances of the three gamma-rays energies involved in  $\delta E$ .

Table 2- Data from Kessler et. al. [5] before the corrections given in the text.

| $E_K$ (keV) | $\sigma_0$ (eV) |
|-------------|-----------------|
| 411.80441   | 0.15            |
| 205.79549   | 0.07            |
| 295.95825   | 0.13            |
| 308.45689   | 0.15            |
| 316.50789   | 0.18            |
| 468.07147   | 0.27            |
| 484.57797   | 0.41            |
| 588.58446   | 0.72            |
| 604.41415   | 0.47            |
| 612.46504   | 0.78            |

Table 3 - Data from Borchert et. al. [6] before the corrections given in the text.

| $E_B$ (keV) | $\sigma_0$ (eV) |
|-------------|-----------------|
| 136.3403    | 0.7             |
| 201.3061    | 0.7             |
| 205.7909    | 0.6             |
| 295.9510    | 0.5             |
| 308.4473    | 2.2             |
| 316.5005    | 0.3             |
| 374.4757    | 0.8             |
| 416.4601    | 3.0             |
| 468.0602    | 0.6             |
| 484.5646    | 1.8             |
| 588.5730    | 4.0             |
| 604.3981    | 2.2             |
| 612.4513    | 2.6             |

Table 4 - Corrected input energies in keV and standard deviations between parentheses, in units of the last figure (first column), and the design matrix  $X$  for the fit, identified by their row numbers.

| $Y_i(\sigma_i)$ | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 | 13 | 14 |
|-----------------|---|---|---|---|---|---|---|---|---|----|----|----|----|----|
| 136.34288(70)   | 1 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0  | 0  | 0  | 0  | 0  |
| 201.30991(71)   | 0 | 1 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0  | 0  | 0  | 0  | 0  |
| 205.79480(61)   | 0 | 0 | 1 | 0 | 0 | 0 | 0 | 0 | 0 | 0  | 0  | 0  | 0  | 0  |
| 295.95661(52)   | 0 | 0 | 0 | 1 | 0 | 0 | 0 | 0 | 0 | 0  | 0  | 0  | 0  | 0  |
| 308.4531(22)    | 0 | 0 | 0 | 0 | 1 | 0 | 0 | 0 | 0 | 0  | 0  | 0  | 0  | 0  |
| 316.50650(33)   | 0 | 0 | 0 | 0 | 0 | 1 | 0 | 0 | 0 | 0  | 0  | 0  | 0  | 0  |
| 374.48279(82)   | 0 | 0 | 0 | 0 | 0 | 0 | 1 | 0 | 0 | 0  | 0  | 0  | 0  | 0  |
| 416.4680(30)    | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 1 | 0 | 0  | 0  | 0  | 0  | 0  |
| 468.06907(64)   | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 1 | 0  | 0  | 0  | 0  | 0  |
| 484.5738(18)    | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 1  | 0  | 0  | 0  | 0  |
| 588.5842(40)    | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0  | 1  | 0  | 0  | 0  |
| 604.4096(22)    | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0  | 0  | 1  | 0  | 0  |
| 612.4629(26)    | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0  | 0  | 0  | 1  | 0  |
| 411.80180(19)   | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0  | 0  | 0  | 0  | 1  |
| 205.79419(9)    | 0 | 0 | 1 | 0 | 0 | 0 | 0 | 0 | 0 | 0  | 0  | 0  | 0  | 0  |
| 295.95638(16)   | 0 | 0 | 0 | 1 | 0 | 0 | 0 | 0 | 0 | 0  | 0  | 0  | 0  | 0  |
| 308.45494(18)   | 0 | 0 | 0 | 0 | 1 | 0 | 0 | 0 | 0 | 0  | 0  | 0  | 0  | 0  |
| 316.50589(20)   | 0 | 0 | 0 | 0 | 0 | 1 | 0 | 0 | 0 | 0  | 0  | 0  | 0  | 0  |
| 468.06851(30)   | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 1 | 0 | 0  | 0  | 0  | 0  | 0  |
| 484.57490(43)   | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 1 | 0  | 0  | 0  | 0  | 0  |
| 588.58073(74)   | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 1  | 0  | 0  | 0  | 0  |
| 604.41032(50)   | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0  | 1  | 0  | 0  | 0  |
| 612.46116(80)   | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0  | 0  | 1  | 0  | 0  |

Table 5. Estimated gamma-ray energies of  $^{192}\text{Ir}$  and  $^{198}\text{Au}$  and standard deviations obtained by the procedure outlined in this paper

| $E_\gamma$ (keV) | $\sigma$ (eV) |
|------------------|---------------|
| 136.34265        | 0.48          |
| 201.30967        | 0.49          |
| 205.79419        | 0.09          |
| 295.95638        | 0.16          |
| 308.45492        | 0.18          |
| 316.50607        | 0.18          |
| 374.48250        | 0.57          |
| 416.46700        | 2.07          |
| 468.06859        | 0.28          |
| 484.57473        | 0.42          |
| 588.58088        | 0.72          |
| 604.41019        | 0.49          |
| 612.46131        | 0.74          |
| 411.80175        | 0.19          |

Table 6 - Covariance (upper triangle, including the main diagonal) and correlation (lower triangle) matrix for the fitted gamma-ray energies. Covariances are in  $\text{eV}^2$ . The rows are labeled by the gamma-ray energies, rounded for the nearest integer, in keV. Lines are in the same order than rows.

|      | 136  | 201  | 206  | 296  | 308  | 317  | 374  | 416  | 468  | 485  | 589  | 604  | 612  | 412 |
|------|------|------|------|------|------|------|------|------|------|------|------|------|------|-----|
| .234 | .029 | .005 | .009 | .008 | .011 | .036 | .116 | .016 | .018 | .024 | .021 | .030 | .008 |     |
| .124 | .237 | .008 | .012 | .012 | .015 | .041 | .119 | .022 | .024 | .031 | .028 | .037 | .014 |     |
| .115 | .167 | .009 | .011 | .011 | .012 | .014 | .016 | .017 | .018 | .022 | .022 | .023 | .015 |     |
| .114 | .159 | .756 | .024 | .016 | .017 | .021 | .028 | .025 | .026 | .032 | .032 | .033 | .022 |     |
| .095 | .136 | .695 | .601 | .031 | .018 | .021 | .025 | .026 | .027 | .033 | .034 | .034 | .023 |     |
| .128 | .171 | .688 | .602 | .547 | .033 | .026 | .038 | .028 | .028 | .035 | .035 | .037 | .025 |     |
| .131 | .147 | .263 | .242 | .212 | .250 | .326 | .143 | .038 | .039 | .050 | .048 | .058 | .029 |     |
| .116 | .119 | .083 | .086 | .070 | .102 | .121 | 4.27 | .056 | .062 | .085 | .073 | .111 | .020 |     |
| .121 | .160 | .657 | .573 | .522 | .535 | .234 | .096 | .080 | .042 | .051 | .052 | .055 | .036 |     |
| .088 | .115 | .458 | .399 | .365 | .371 | .164 | .072 | .353 | .176 | .053 | .054 | .057 | .035 |     |
| .069 | .087 | .324 | .283 | .258 | .264 | .121 | .057 | .251 | .177 | .520 | .066 | .070 | .042 |     |
| .090 | .118 | .490 | .427 | .390 | .395 | .171 | .072 | .376 | .263 | .188 | .239 | .070 | .044 |     |
| .084 | .103 | .327 | .288 | .261 | .272 | .137 | .072 | .259 | .182 | .131 | .193 | .555 | .044 |     |
| .088 | .149 | .846 | .735 | .670 | .701 | .264 | .050 | .653 | .439 | .306 | .470 | .309 | .037 |     |

Table 7 - Cascade/cross-over energies differences between gamma-rays from  $^{192}\text{Ir}$  decay. Same as table 1, with the data from tables 5 and 6. The total  $\chi^2$  equals 2.6.

| $E_1$ (keV) | $E_2$ (keV) | $E_{\text{co}}$ (keV) | $\delta E$ (eV) <sup>a</sup> | $\sigma_E$ (eV) <sup>b</sup> |
|-------------|-------------|-----------------------|------------------------------|------------------------------|
| 136.34265   | 468.06859   | 604.41019             | 0.69                         | 0.66                         |
| 295.95638   | 308.45492   | 604.41019             | 0.60                         | 0.44                         |
| 295.95638   | 316.50607   | 612.46131             | 0.62                         | 0.71                         |

a) Energy differences corrected for recoil energies

b) Covariance matrix taken into account. Forgetting the covariances, this column would be, in the order of appearance on the table: 0.74, 0.54, and 0.78, respectively.

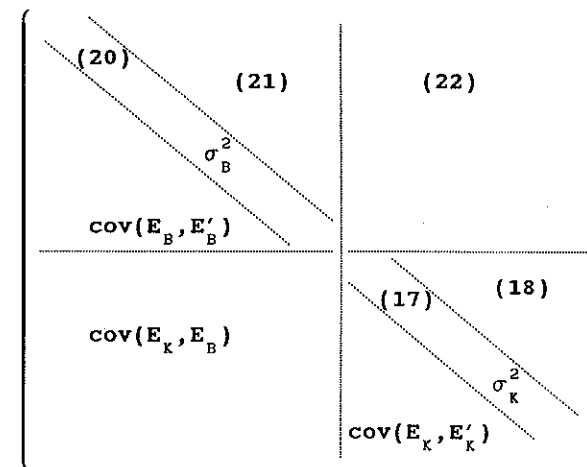


Figure 1 - Diagrammatic representation of the fit covariance matrix. The upper-left square contains the covariances between data from [6], the lower-right square contains the covariances between data from [5], and the upper-right and lower-left rectangles contain the covariances between data from [6] and [5]. Enclosed in parentheses are the numbers of the corresponding covariance formulas in the text. The matrix is symmetric and the main diagonal, which contains the variances, is explicitly shown.