Determination of207Bi activity and detection efficiencies by using the method of X-gamma coincidences

Authors: Nađđerđ, Dr. Laslo, Jordanov, Dr. Dragana, Inoyatov, Dr. Anvar, Khushvaktov, Dr. Jurabek, Kolesnikov, Dr. Vadim, Devetak, Dr. Damir, Milošević, Dr. Jovan, Nađđerđ, Dr. Laslo

Date: 2026-05-14T21:56:51+00:00

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Full Text

Preamble

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Devetak1,2, and J. Milošević1,2 Vinča Institute of Nuclear Sciences - National Institute of the Republic of Serbia, University of Belgrade, Mike Petrovića Alasa 12-14, Vinča 11351, Belgrade, Serbia.

Strong coupling Physics International Research Laboratory, Huzhou University, Huzhou, Zhejiang 313000, P. R. China.

Joint Institute for Nuclear Research (JINR), Dubna, Russian Federation.

Abstract

We present an enhancement of the matrix method of X-gamma coincidences by simplifying the way of obtaining counting rate equations of the commonly used detection pathways. That empowers getting an algebraically non-truncated, closed-form, counting rate equations for the complex schemes of radioactive decays. We applied the method to the radioisotope of bismuth, 207Bi. The daughter nuclei of this radioisotope have a metastable state that induces the splitting of the true coincidence pathways which have to be addressed in the counting rate equations.

Keywords

Gamma spectrometry, Coincidence summing, Isomeric transition, Bismuth 207Bi

Introduction

The true coincidence summing in the energy spectra of photons, emitted at cascade deexcitations of radionuclide progenies, is readily used for standardization of radioactive sources [1-5]. Semkow et al. [6] founded the matrix formalism for handling true coincidencesumming effects, although their approach did not consider cases in which X-photons participate in the summing. The complete matrix method of X-gamma coincidences is presented in Ref. [7]. The method describes formation of the counting rate equations for all possible X-gamma detection pathways for a radioactive decay. In Fig. 1 [Figure 1: see original paper] is shown a scheme of radioactive decay, where the ground state of the parent nucleus is presented as the highest excited state m of the daughter nucleus in order to adapt it to the matrix formalism.

In the present work we slightly modify the original notation in order to make the method clearer and easier to implement. Namely, the detecting probability matrices for X (Kα, K and photons are given by A, B, and Γ respectively, in addition to a non-detecting probability matrix N:

ê 1, 0 A = ê a2,0 êam , 0

a 2,1

ê 1, 0 Γ = ê 2, 0

ê 1, 0 0ú, B = ê b2, 0 êbm , 0

0 0ú ê 1, 0 ú ê nm , 0 0 0úû

2,1

bm ,1 bm , m -1

nm ,1 nm , m -1

Elements in the last rows of these matrices are the detecting probabilities in the electron capture transitions. For example, the probabilities of detecting Kα and K photons, along with the probability of non-detecting any X-photon, are given respectively by: am,i = xm,ie a g a wK PKi

bm,i = xm,ie g wK PKi

nm,i = xm,i [1 - (t a g a + t g)wK PKi] where xm,i are the corresponding transition probabilities, ea and e are the full-energy peak efficiencies, ta and t are the total efficiencies of detection of Kα and K photons respectively; ga and g are their corresponding fractions, w is the K-shell fluorescent yield, and PKi are the probabilities of capture of the K-shell electron and the consequent transition of the parent nucleus to the ith level of the daughter nucleus. However, if one deals with + - decays, instead of the electron-capture ones, the matrix elements am,i and bm,i are equal to zero and qm,i are reduced to the transition probabilities xm,i.

All other elements of the detecting probability matrices (1) correspond to the internal conversion transitions. For example, probabilities of detecting X (Kα and K), and photons, and non-detecting any photon, are given respectively by:

a iK, j ai,j = xi , je g wK 1 + a iT, j

a iK, j bi,j = xi , je g wK 1 + a iT, j

i,j = x e 1 + a iT, j i, j i, j

a iK, j ù ni,j = xi , j ê1 - ti , j - (t g + t g)wK 1 + a iT, j 1 + a iT, j úû where a i,Kj are the K-shell internal conversion coefficients, a i,T j are the total conversion coefficients, e i, j are the full-energy peak (FEP) efficiencies and t i , j are the total efficiencies of detection of gamma photons emitted at the quoted transitions. We kept the transition

indices in the notation for gamma detection efficiencies because nuclear de-excitations are followed by emission of photons of different energies unlike the X photons involved in the coincidence summing which have all equal energies for all nuclear transitions, the electron capture, and the internal conversion ones.

In the next Section we introduce an enhancement to the original method. The application of the method on the radioisotope of bismuth 207Bi is presented in Section 3, and the experimental verification of the method is presented in Section 4.

Enhancement of the method

The original exposition of the matrix method of X-gamma coincidences proceeds with a creation of the total detecting probability matrix followed by the extraction of all detecting outcomes [7]. However, one is often not interested into the algebra of separation of the counting rate equations, but rather to write them exactly in a straightforward way in order to find their exact solutions. Here we expose a shortcut to some of the most used equations of the method. For example, the total counting rate equation might be simple written as following:

I (ETot) = [1 - M m,0]R ,

where R is the activity of the radioactive source, and M is a matrix obtained by summing up the powers of the non-detection probability matrix N:

M = å Nk .

Similarly, a counting rate equation of a gamma spectral line detected in an energy spectrum, regardless of the number of gamma-gamma coincidences involved, is given by the following formula:

ìM m,iG i , j M j ,0 R if M m,iG i ,0 R if

where G is a matrix obtained by summing up the powers of the gamma-detection probability matrix Γ :

G = å Γk .

However, in the case of coincident detection of the non-consecutive gamma transitions, k→i, and j→l, the count rate equation contains another matrix element, M i, j which connects the ending level i of the first transition with the starting level j of the succeeding transition:

ìM m, k G k ,i Mi , j G j ,l Ml ,0 R if l 1 0 î M m, k G k ,i Mi , j G j ,0 R if l = 0

and so on for higher number of non-consecutive gamma transitions.

All these equations, given in (6) and (8), start with a non-detection probability matrix M from the ground state of the parent nucleus, m, because the transitions from this level are never followed by gamma de-excitations.

The count rate equation of the X (Ka) spectral line is given by the following formula: m -1 j -1 j = 2 i =1

where the first two terms, led by elements of matrix A , count the contribution of detecting X-photons emitted at the electron-capture decays. On the other hand, the last two terms count the contribution of detecting X-photons which come from the internal conversion, depending on whether the emission of the Ka photon occurs at the final step of the transition, or not.

Using the given recipe one can construct a complete count rate equation for any X-gamma coincident sum-peak. For example, the case of true coincidence of Ka photons and gamma photons emitted at the de-excitation from the first excited state: m -1 j -1 I (EKa + E1,0) = ê A m,1 + å A m,i M i ,1 + å M m,i Ai ,1 + åå M m, j A j ,i M i ,1 úG1,0 R . j =3 i = 2

Another interesting case for observation is the count rate equation for the Ka-Ka coincidence which is often used in the X-gamma coincidence method for calculation of the detection efficiencies: m -1 i -1 m -1 i -1 é m -1 I (EKa +

EKa) = êå A m,i Ai ,0 + åå A m,i Ai , j M j ,0 + åå A m,i M i , j A j ,0 + i = 2 j =1 i = 2 j =1 ë i =1 m -1 i -1 j -1

m -1 i -1

- ååå A m,i M i , j A j , k M k ,0 + åå M m,i Ai , j A j ,0 +

i = 2 j =1

m -1 i -1 j -1

m -1 i -1 j -1

- ååå M m,i Ai , j A j , k M k ,0 +ååå M m,i Ai , j M j , k A k ,0 +

m -1 i -1 j -1 k -1 + åååå M m,i Ai , j M j , k A k ,l M l ,0 ú R

Here, the first four sums contain the electron-capture contributions, because they all start with the Mm,i detection probability, and the last four ones are the pure internal conversion sums. It is easy to observe a pattern present in Eq. (11). Namely, all terms contain two A matrix elements which occupy all distinct positions in the multiple sums. Their indices, on the other hand, just follow the rule of successive transitions up to the final ground level of the daughter nuclei.

Unlike the original approach of the matrix method of X-gamma coincidences, where the counting rate equations for complex decay schemes are usually truncated due to enormous number of terms, such those shown in Refs. [8, 9], the equations given in the present section

are complete count rate equations, convenient for numerical calculations.

Application to 207Bi

Bismuth 207Bi decays by electron capture to the excited nuclear states of 207Pb, shown in a modified scheme in Fig. 2 [Figure 2: see original paper], although a week + transition to the first excited state of 207Pb has also been revealed [10].

Applying the matrix method of X-gamma coincidences in its enhanced form, illustrated in the previous section, allows an easy construction of all counting rate equations for the radioisotope of bismuth 207Bi, as shown in Eq. (12). These equations are composed of the FEP, and the total detection efficiencies by means of Eqs. (1-3). Therefore, if the detection efficiencies are the unknowns, it is easy to construct a closed-form system of non-linear equations for their determination. These equations are already written in a form convenient for numerical calculations:

I (ETot) = [1 - M 5, 0]R 4 j -1 j = 2 i =1 4 j -1 j = 2 i =1 I (E2,1) = M 5, 2G 2,1M1, 0 R

I (E1, 0) = M 5,1G1, 0 R 4 j -1 j =3 i = 2

I (E3,1) = M 5,3G 3,1M1, 0 R I (E3, 0) = M 5,3G 3, 0 R I (E4,1) = M 5, 4G 4,1M1, 0 R I (E4, 2 + E1, 0) = M 5, 4G 4, 2 M 2,1G1, 0 R

In a high-resolution energy spectrum of this radioisotope, shown in Fig. 4 [Figure 4: see original paper], Ka1 and Ka appear as separate peaks, as well as the K 1 and K , which justify the formation of separate counting rate equations for each of the recorded peaks. To achieve this objective, we assigned new detecting probability matrices C and D , with elements ci,j and di,j, which correspond to the Ka and K   peaks, respectively, with the corresponding fractions ga and g , given in the tables of radioisotopes [10]. If one prefers the expanded form of equations, over those given

by Eq. (12), with an explicit notation using the detecting probabilities, they are given by the following equations:

I (ETot) = [1 - n54 n42 n20 - (n54 n42 n21 + n54 n41 + n53n31 + n51)n10]R I (EKa 2) º I (72.8) = (n54c42 n20 + n54 n42c20 + n54c42 n21n10 + n54 n42 n21c10 + n54c41n10 + + n54 n41c10 + c53n31n10 + n53c31n10 + n53n31c10 + c51n10 + n51c10) R I (EKa 1) º I (75.0) = (n54 a42 n20 + n54 n42 a20 + n54 a42 n21n10 + n54 n42 n21a10 + n54 a41n10 + + n54 n41a10 + a53n31n10 + n53a31n10 + n53n31a10 + a51n10 + n51a10) R I (EK 1) º I (84.9) = (n54b42 n20 + n54 n42b20 + n54b42 n21n10 + n54 n42 n21b10 + n54b41n10 + + n54 n41b10 + b53n31n10 + n53b31n10 + n53n31b10 + b51n10 + n51b10) R I (EK 2) º I (87.2) = (n54 d 42 n20 + n54 n42 d 20 + n54 d 42 n21n10 + n54 n42 n21d10 + n54 d 41n10 + + n54 n41d10 + d53n31n10 + n53d31n10 + n53n31d10 + d51n10 + n51d10) R

I (E2,1) º I (328.1) = n54 n42 21n10 R I (E1, 0) º I (569.7) = (n54 n42 n21 + n54 n41 + n53n31 + n51) 10 R I (EKa 1 + E1, 0) º I (644.7) = (n54 a42 n21 + n54 a41 + n53a31 + a51) 10 R I (E2, 0) º I (897.8) = n54 n42 (20 +   21 10) R I (E3,1) º I (1063.7) = n53 31n10 R I (E4, 2) º I (1442.2) = n54 42 (n20 + n21n10) R I (E3, 0) º I (1633.4) = n53 31 10 R I (E4,1) º I (1770.2) = n54 (42 21 +   41)n10 R

I (E4, 2 + E1, 0) º I (2011.9) = n54 42 n21 10 R I (E4, 0) º I (2339.9) = n54 (42 21 10 +   42 20 +   41 10) R

Nevertheless, from the standpoint of X-gamma coincidences 207Bi has an interesting feature which we found challenging to be analyzed. Namely, the third excited state of its daughter nuclei is a metastable state, shown in Fig. 2, which lasts ~800 ms, which is much longer than the detectors' charge collection time. This means that X photons, emitted at the electron capture transitions to the metastable state, cannot be detected in true coincidence with any other photons. Also, the photons emitted in cascade transitions from the metastable state cannot be in true coincidence with any other photons. To make this statement clearer we made a scheme of time-separated coincident transitions, shown in Fig. 3 [Figure 3: see original paper].

There are three separate groups of transitions, marked by green ellipses in Fig. 3. Photons emitted in transitions in each of these groups might be detected in true coincidences.

However, there can never be true coincidences between transitions belonging to distinct groups. In another words, there are no mixing between these three groups of transitions. This peculiarity has a certain implications on the counting rate equations. Namely, each probability of non-detecting of X-photons n5,3, marked by red rectangles in Eq. (13), has to be replaced by the corresponding transition matrix x5,3 to take into account the influence of the metastable state: n53 ® x53 .

The requirement of non-detection of the X-photon in this transition, by using n5,3, is too restrictive because even if the photon is detected, that event would not contribute to coincidence summing due to the isomerism of the third excited state. Thus, in all indicated points in Eq. (13), where the replacement (14) should be applied, only the transition has to happen in order to start the corresponding cascade. The replacement is not to be done only in the first equation for the total counting rate, because there one needs to fully address the nondetecting probability role.

By using the same arguments one needs also the following replacements to fully address the impact of the mentioned isomerism:

a53n31n10 ® a53 , b53n31n10 ® b53 , c53n31n10 ® c53 , d 53n31n10 ® d 53 .

Namely, all cascades which start with the probabilities of detection of any X-photon, through the metastable state, does not require any non-detection because it will eventually end up in the ground state, and the detecting outcome cannot be in true coincidence with any of the subsequent event of detection.

The calculation results were tested by comparison with experimental data from the FEP detector efficiencies for gamma lines from the decay of 207Bi.

Experiment

A commercial 207Bi source from a set of special spectrometric gamma sources (SSGS) manufactured by RITVERTS Joint-Stock Company (Russia) was used for the measurements.

The activity of the 207Bi source at the time of measurement was 5.30(16) kBq. The activities of impurity gamma-emitting radionuclides were $\leq$ 0.1% of the 207Bi activity. The size of the active part of the source was Ø ~2.5 mm.

The spectra of X- and gamma-rays emitted by the source were measured using a Canberra GR 1819 HPGe detector with a 0.5 mm thick beryllium window. The relative efficiency of the detector according to the specification is 18.9%. The energy resolution at the energy 122 keV gamma line (57Co) is 0.8 keV. Sources from the SSGS suite were used to determine the absolute efficiency and energy calibration of the detector in the energy range from 30 to 2400 keV. The characteristic measurement time of the 207Bi source and calibration sources was ~3 days. The dead time of the registration in the measurements was below 2%. A measurement time of one spectrum was usually 3 days. The distance between source and detector in all measurements was 1 cm. The measured spectra were evaluated by computer code DEIMOS described in the work [11]. The result of a measurement is shown in Fig. 4 where the gamma full-energy peaks of 207Bi are well observable along with the corresponding X-gamma, and gamma-gamma true coincidence peaks. Even a single (SE) and double (DE) escape peaks of the peak I(E4,1) might be seen.

By solving the appropriate system of non-linear equations (12) we obtained the FEP detection efficiencies at 207Bi energies shown by closed red stars in Fig.5. In order to compare the results we measured the standard sources of 139Ce, 133Ba, and 152Eu under the same experimental conditions. The certified values of their activities were 38.9 Bq, 3728 Bq, and

850 Bq, respectively, with 3% uncertainties given on the confidence level of 95%. Applying the same method of X-gamma coincidences to the standard sources we obtained the fullenergy peak efficiencies of the detector in a broad energy range, shown in Fig. 5 [Figure 5: see original paper]. The statistical uncertainties of the efficiencies, driven by the measuring uncertainties of the counting rates, are shown by vertical bars. The total uncertainties, which also include the uncertainties of the nuclear and atomic parameters, are shown by wide gray vertical bars. The results for 207Bi energies fit well with the ones obtained by using the standard source. Also, by treating activities of the sources as unknowns we obtained results that overlap with the certified ones.

Summary and conclusion

We presented an enhancement of the matrix method of X-gamma coincidences that empowers getting an algebraically non-truncated, closed-form, counting rate equations for the complex schemes of radioactive decays. The method is applied to the radioisotope of bismuth, Bi which progeny have a metastable state. By taking into account the presence of the metastable state we obtained the full-energy peak efficiencies of a detector at 207Bi energies.

The correctness of the approach was confirmed by comparison of the results by those obtained by using standard radionuclide sources.

Acknowledgments

The authors acknowledge the support from Ministry of Science, Technological Development and Innovation of Republic of Serbia throughout the theme 0102602, Collaboration Projects JINR-Serbia_P23, and JINR-Serbia_P211.

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Note: Figure translations are in progress. See original paper for figures.