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THE NEW METHOD FOR SOLVING GAMMA-GAMMA, X-X, X-GAMMA COINCIDENCE SUMMING IN GAMMA SPECTROSCOPY

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Abstract: *New challenges in gamma spectroscopy came with the development of experimental techniques. The new germanium detectors have an extended detector sensitivity to lower energies - BEGe (broad energy HPGe), which allows the efficient detection of X-rays generated in the processes of electronic capture and internal conversion at the same time as gamma radiation. In addition to the previous gamma-gamma, this type of detector has an additional X-X and X-gamma coincidence summation, which further complicates the problem of deconvolution of the energy spectrum. By considering and analyzing previously developed methods, a deterministic method was developed for deriving the counting rate for gamma spectra, which allows us to predict all summation peaks that occur in the spectrum. We devised a method for calculating coincident effects in gamma spectroscopy that is applicable to all radionuclides regardless of the degree of complexity of the decay scheme. This approach also allows us to determine the activity of sources directly without calibration of the detector, which is very important in metrology of radionuclides. Accordingly, based on the value of the peak area in the spectrum and knowledge of probability transitions between excited states of a nucleus, it is possible to determine the efficiency of detection together with the activity of radioactive sources. Our work also includes the successful application of the new method to radionuclides used in real experiments such as radionuclides* ^{139}Ce, ^{57}Co, ^{133}Ba *and* ^{152}Eu.

Keywords: *Metrology of radionuclides, Coincidence summing, Activity of source, Detector efficiency,* ^{152}Eu.

1. INTRODUCTION

Coincidental summation is topical in international groups dealing with radionuclide metrology, i.e. gamma radiation spectrometry.

Interest in the effects of coincidental summation in gamma spectrometry is related to their influence on the accuracy of determining the efficiency of detectors and radionuclide activity.

Our goal was to find the most optimal method for calculating the effects of coincidental summation in gamma spectroscopy and we developed a new matrix method for calculating the counting rate for radionuclides in which the cascade deexcitation of the nucleus takes place simultaneously with the cascade deexcitation of the atomic shell [1-4].

Applying the analytical approach gives the problem of coincidental summation the ability to predict all the summation peaks that may appear in the spectrum.

The new method has been successfully applied to radionuclides used in real metrology. We successfully applied the new matrix method to radionuclides with a simple decay scheme. It was also applied with the same success to the radionuclide with an extremely complex decay scheme ^{152}Eu [2].

When we were working on ^{152}Eu, using a new method for calculating the effects of coincidental summation, certain irregularities were noticed during the formation of probability matrices. Analyzing the calculations given in the data evaluation, which follow the tables with transition probabilities, a series of errors and omissions are observed that directly affect the values of the recommended transition intensities.

To facilitate future work in gamma spectroscopy we give in the paper, an analysis of the errors and mistakes that the evaluators have made [3].

2.THE NEW MATRIX METHOD

2.1.The matrix method for calculating coincident effects

During nuclear decay, cascading transitions emit two or more photons, in a short time interval, shorter than the resolving time of the detector, and as a result, the detector treats them as a single interaction. This leads to an

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increase in counts in the summation peaks as well as a decrease in the counts in the photo peaks of the cascading photons (Figure 1.). Andreev et al.[5] have method were determines nuclear parameters, while source activities and detection efficiencies are known. We developed a method where we solve the inverse problem: knowing the nuclear parameters, we determine the unknown activities of the source and the efficiency of detection.

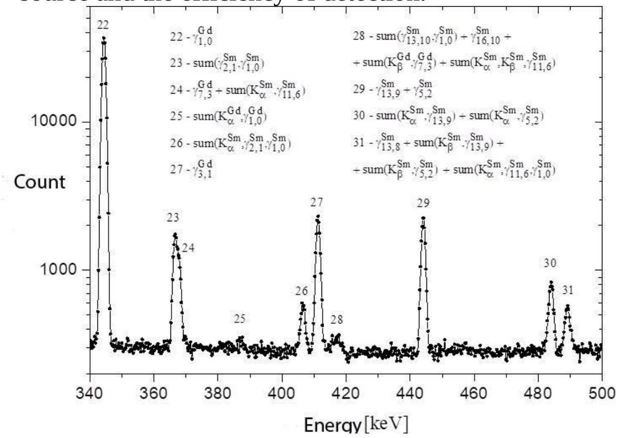


Figure 1. The part of the spectrum of Eu-152 where the summation peaks are shown

That is, based on the values of the peak areas in the spectrum and knowing the probabilities of transitions between the excited nucleus states, we determine the activity and efficiency. First, we modified the decay scheme for the needs of matrix formalism (Figure 2.)

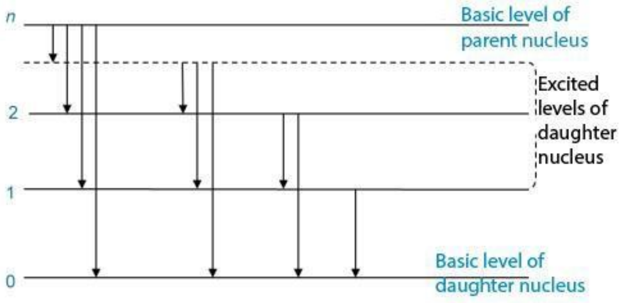


Figure 2. The decay scheme is modified as follows: the ground level of the parent nucleus is represented as the most excited level of the daughter nucleus and is denoted by *n*

Then we form the transition probability matrix (stochastic, transition or probability matrix) X: transition probabilities x_{ij} from the energy level i to the j level form one square matrix of dimensions (n + 1) × (n + 1)

$$\mathbf{X} = \begin{bmatrix} 0 & 0 & 0 & \cdots & 0 \\ x_{10} & 0 & 0 & \cdots & 0 \\ x_{20} & x_{21} & 0 & \cdots & 0 \\ \vdots & \vdots & \vdots & \vdots & \vdots \\ x_{n0} & x_{n1} & \cdots & x_{nn-1} & 0 \end{bmatrix} \tag{1}$$

$$\sum_{i=0}^{n-1} x_{ni} = 1 \tag{2}$$

$$\sum_{j=0}^{i-1} x_{ij} = 1, \quad i = 1, \ldots, n-1 \tag{3}$$

When the nucleus's decay occurs near the detector, the photons emitted in the cascade transitions can be detected. The set of all detection probabilities forms a characteristic diagram. However, it is necessary to come to an expression for the probabilities of simultaneous detection of photons during the cascade transition, i.e. the transition in several successive steps. In fact, it should only be noted that the expressions for detection probabilities are nothing but expressions for matrix elements of matrices that we have called detection probability matrices:

- Probability matrix of K_α photon detection, A,
- Probability matrix of K_β photon detection, B,
- Probability matrix of gamma photon detection, Γ,
- Non-detection probability matrix of any photon, Q.

Summation matrix of all detection probability matrices:

$$S=A+B+\Gamma+Q, \tag{4}$$

contains all probabilities of (non) detection of photons emitted during one-step transitions.

Analogously, the sum of all powers of the matrix S contains all the detection probabilities of photons emitted at cascade transitions in any possible number of steps:

$$\mathbf{M} = \sum_{k=1}^{\nu-1} \mathbf{S}^k = \mathbf{S} + \mathbf{S}^2 + \mathbf{S}^3 + \ldots + \mathbf{S}^{\nu-1} \tag{5}$$

For the problem of coincidental summation, only complete cascade transitions are interesting, i.e. cascade transitions that start from the basic state of the parent nucleus and end with the basic state of the daughter nucleus. Therefore, instead of the whole matrix M, which contains all possible detection probabilities, for our further analysis we need only its element which is in the last, n-th row, and zero column:

$$
\begin{aligned}
[\mathbf{M}]_{n,0} = {} & \overbrace{a_{n,0}e^{iE_\alpha} + b_{n,0}e^{iE_\beta} + q_{n,0}e^{i0}}^{\text{1 корак}} + \\
& + \overbrace{\left(a_{n,n-1}e^{iE_\alpha}a_{n-1,0}e^{iE_\alpha} + a_{n,n-2}e^{iE_\alpha}a_{n-2,0}e^{iE_\alpha} + \dots + a_{n,1}e^{iE_\alpha}a_{1,0}e^{iE_\alpha}\right)}^{\text{2 корака}} + \\
& + \left(a_{n,n-1}e^{iE_\alpha}b_{n-1,0}e^{iE_\beta} + a_{n,n-2}e^{iE_\alpha}b_{n-2,0}e^{iE_\beta} + \dots + a_{n,1}e^{iE_\alpha}b_{1,0}e^{iE_\beta}\right) + \\
& + \left(a_{n,n-1}e^{iE_\alpha}\gamma_{n-1,0}e^{iE^{\gamma}_{n-1,0}} + a_{n,n-2}e^{iE_\alpha}\gamma_{n-2,0}e^{iE^{\gamma}_{n-2,0}} + \dots + a_{n,1}e^{iE_\alpha}\gamma_{1,0}e^{iE^{\gamma}_{1,0}}\right) \\
& + \left(a_{n,n-1}e^{iE_\alpha}q_{n-1,0}e^{i0} + a_{n,n-2}e^{iE_\alpha}q_{n-2,0}e^{i0} + \dots + a_{n,1}e^{iE_\alpha}q_{1,0}e^{i0}\right) + \\
& + \left(b_{n,n-1}e^{iE_\beta}a_{n-1,0}e^{iE_\alpha} + b_{n,n-2}e^{iE_\beta}a_{n-2,0}e^{iE_\alpha} + \dots + b_{n,1}e^{iE_\beta}a_{1,0}e^{iE_\alpha}\right) + \\
& + \left(b_{n,n-1}e^{iE_\beta}b_{n-1,0}e^{iE_\beta} + b_{n,n-2}e^{iE_\beta}b_{n-2,0}e^{iE_\beta} + \dots + b_{n,1}e^{iE_\beta}b_{1,0}e^{iE_\beta}\right) + \\
& + \left(b_{n,n-1}e^{iE_\beta}\gamma_{n-1,0}e^{iE^{\gamma}_{n-1,0}} + b_{n,n-2}e^{iE_\beta}\gamma_{n-2,0}e^{iE^{\gamma}_{n-2,0}} + \dots + b_{n,1}e^{iE_\beta}\gamma_{1,0}e^{iE^{\gamma}_{1,0}}\right) \\
& + \left(b_{n,n-1}e^{iE_\beta}q_{n-1,0}e^{i0} + b_{n,n-2}e^{iE_\beta}q_{n-2,0}e^{i0} + \dots + b_{n,1}e^{iE_\beta}q_{1,0}e^{i0}\right) + \\
& + \left(q_{n,n-1}e^{i0}a_{n-1,0}e^{iE_\alpha} + q_{n,n-2}e^{i0}a_{n-2,0}e^{iE_\alpha} + \dots + q_{n,1}e^{i0}a_{1,0}e^{iE_\alpha}\right) + \\
& + \left(q_{n,n-1}e^{i0}b_{n-1,0}e^{iE_\beta} + q_{n,n-2}e^{i0}b_{n-2,0}e^{iE_\beta} + \dots + q_{n,1}e^{i0}b_{1,0}e^{iE_\beta}\right) + \\
& + \left(q_{n,n-1}e^{i0}\gamma_{n-1,0}e^{iE^{\gamma}_{n-1,0}} + q_{n,n-2}e^{i0}\gamma_{n-2,0}e^{iE^{\gamma}_{n-2,0}} + \dots + q_{n,1}e^{i0}\gamma_{1,0}e^{iE^{\gamma}_{1,0}}\right) + \\
& + \left(q_{n,n-1}e^{i0}q_{n-1,0}e^{i0} + q_{n,n-2}e^{i0}q_{n-2,0}e^{i0} + \dots + q_{n,1}e^{i0}q_{1,0}e^{i0}\right) + \\
& + \overbrace{\left(a_{n,n-1}e^{iE_\alpha}a_{n-1,n-2}e^{iE_\alpha}a_{n-2,0}e^{iE_\alpha} + \dots + a_{n,2}e^{iE_\alpha}a_{2,1}e^{iE_\alpha}a_{1,0}e^{iE_\alpha}\right)}^{\text{3 корака}} + \\
& + \left(a_{n,n-1}e^{iE_\alpha}a_{n-1,n-2}e^{iE_\alpha}b_{n-2,0}e^{iE_\beta} + \dots + a_{n,2}e^{iE_\alpha}a_{2,1}e^{iE_\alpha}b_{1,0}e^{iE_\beta}\right) + \\
& + \cdots + \\
& + \underbrace{q_{n,n-1}e^{i0}q_{n-1,n-2}e^{i0}q_{n-2,n-3}e^{i0}\cdots q_{1,0}e^{i0}}_{\nu-1\ \text{корака}}
\end{aligned}
\tag{6}
$$

Different detection events whose probabilities in the previous expression have the same argument actually belong to a group of events with the same detection outcome. All of these members contribute to the same photo peak. This finally established a connection with the energy spectrum. The normalized area below a certain peak in the energy spectrum is equal to the sum of the probabilities of detection of all events with the same outcome.

The theoretical counting rate in the corresponding photo-peak is obtained by multiplying the activity of the source, R, with the sum of the probabilities of detection of all events with the same outcome:

$$n^{\mathrm{Th}}(E_\ell) \equiv R\sum_k p_k(E_\ell) \qquad \ell = 1,2,3,\dots \tag{7}$$

By equating theoretical with experimental counting rate, a system of equations for counting rate in all peaks in the energy spectrum is obtained:

$$R\sum_k p_k(E_\ell) = n^{\mathrm{Exp}}(E_\ell) \qquad \ell = 1,2,3,\dots \tag{8}$$

that is, a system of equations based on when we determine R and all efficiencies.

3.EXPERIMENT

3.1Equipment

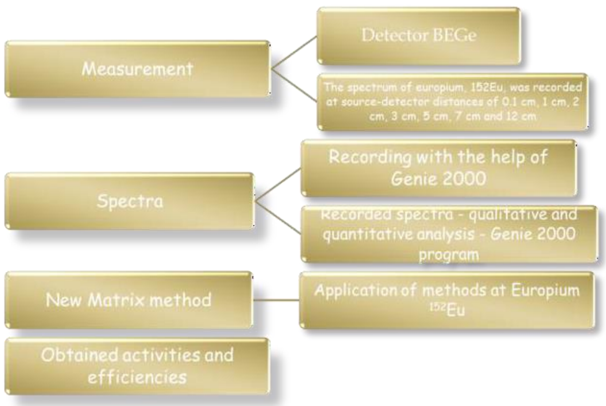
One of the important characteristics of detectors for gamma radiation detection is the simultaneous detection of photons (emitted in the process of cascade deexcitation) which lead to the appearance of summation peaks (coincidental summation). Previously detectors did not detect X-rays or were removed by filters. New challenges in gamma spectroscopy came with the development of experimental techniques. The new germanium detectors have an extended detector sensitivity to lower energies - BEGe (broad energy HPGe), which allows the efficient detection of X-rays generated in electronic capture and internal conversion processes at the same time as gamma radiation (Figure 3.).

Figure 3. Extendent Range Coaxial Canberra Ge detector GX5020

Scientists have successfully developed a method for solving gamma-gamma coincidence summation. But in BEGe - in addition to the previous gamma-gamma, this type of detector has an additional X-X and X-gamma coincidence summation, which further complicates the problem of deconvolution of the energy spectrum.

3.2.Experimental settings

We verified our matrix method using experimental spectra.



Point source of europium, ^{152}Eu, reference activity 7037 Bq ± 4% on the day of calibration on June 9, 1987, measured on September 27, 2011. a high-purity germanium detector with an energy range extended to lower values (Extendent Range Coaxial Canberra Ge detector GX5020. The extended energy range of this type of detector enables efficient detection of Kα and Kβ radiation. The dead time of measurement in the nearest geometry is below 3%.

After subtracting the background, the spectrum of Europium ^{152}Eu was analyzed using the Genie2000 program. The total number and area under all peaks together with their errors were determined.

The results of calculated values of activities from the experimental spectra are shown in **Table 1**. We have a good agreement with value of activity point source from Reference Laboratory

Table 1. Results of measurement and calculation

Referent activity	2023 ± 81 Bq						
Spectrum number	1	2	3	4	5	6	7
The distance of the source from the detector	*0.1cm*	*1cm*	*2cm*	*3cm*	*5cm*	*7cm*	*12cm*
Calculated value of source activity	*2070 ± 26 Bq*	*2083 ± 34 Bq*	*2076 ± 35 Bq*	*2142 ± 54 Bq*	*2088 ± 127 Bq*	*2169 ± 84 Bq*	*1882 ± 109 Bq*

4.CONCLUSION

With the development of experimental techniques in gamma spectroscopy, the appearance of more sensitive detectors, which can measure even lower energies, and therefore, in addition to gamma radiation and X-ray radiation, it became necessary to develop a method for calculating the coincidental effects that arise.

We have developed a method that allows us to determine an additional X-X and X-gamma coincidence summation in addition to the previous gamma-gamma. Using the data given in the transition probabilities tables, we formed, following our method, the appropriate detection probability matrix. This approach allows us to determine activity of source and efficiency of detection. The importance of the new method is that it allows us to determine the efficiency of detection and activity of radioactive sources directly without calibration of the detector, which is very important in the metrology of radionuclides.

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