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Two-dimensional diagonal summing of coincidence spectra for bulk PGNAA applications

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Abstract

In the past 10 years, new electronic devices have been developed that allow fast coincidence measurements to be performed that are capable of simultaneously recording the individual spectra as well as the coincidence spectra of multiple detectors. Utilizing these devices with computer software allows multiparameter data acquisition which adds much more flexibility in data analysis. One of the capabilities that is enabled is that of obtaining two-dimensional spectra.

In this work, the use of this equipment and the two-dimensional spectra obtained with it are used to allow two-dimensional diagonal summing. The main advantages of this approach are improved peak resolution and very low background (Compton continuum). Possible uses of the two-dimensional diagonal summing are identifying coincidence schemes, performing elemental analysis, and identifying trace elements in bulk samples. The spectra obtained are very promising for these applications.

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1. Introduction

One of the main advantages of γ–γ coincidence counting is the elimination of the background spectrum, pulse pile-up, and summing effects (for simple schemes). For prompt gamma-ray neutron activation analysis (PGNAA), the sources of background include the gamma rays from the natural background, from surrounding materials, from the neutron source, and from detector neutron activation. Feasibility studies for PGNAA [1] for the γ–γ coincidence counting approach were previously conducted. These studies demonstrated the elimination of the non-coincident background spectrum and an increase in the Signal-to-Noise (S/N) ratio.

In the past 10 years, new electronic devices have been developed that allow fast coincidence measurements to be performed that are capable of simultaneously recording the individual spectra as well as the coincidence spectra of multiple detectors. Utilizing these devices with computer software allows multiparameter data acquisition which adds much more flexibility in data analysis.

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One of the capabilities that is enabled is that of obtaining two-dimensional spectra. The values in the two-dimensional spectra represent the counts corresponding to the different channel pairs in the detectors used in the coincidence experiment. These types of spectra provide us with more detailed coincidence information than that from one-dimensional spectra [2].

In this work, the two-dimensional spectra are presented in a flat view where the coincidence intensity is defined by the darker areas in the plot. Extracting part (or all) of the information from the flat view plots can transform them into normal one-dimensional plots. This is normally done by summing (projecting) a certain area in the two-dimensional plot along one of the axes. This projection can be made in four different ways:

1. Projecting the whole spectrum area to one of the axes. This yields the total coincidence spectrum in one of the detectors. This is the same as summing all the rows (or columns) in the coincidence data array.
2. Projecting a horizontal (or vertical) range of the spectrum to one of the axes. This is called horizontal (or vertical) window summing. On one of the axes this would represent an energy range equivalent to the width of the window. On the other axis this would represent the spectrum that is in coincidence with this energy range.
3. Projecting a diagonal area of the spectrum to one of the axes. This is the main focus of this work and will be explained in further detail in the following section.
4. Projecting an irregular range of the spectrum to one of the axes. This type of projection depends on the type of physical problem being considered. A typical example of this is recording the pulse amplitude and rise time in coincidence to identify the source of the pulse, whether it is alphas, betas, or gammas [3].

2. Two-dimensional diagonal summing

Projecting a diagonal area in a two-dimensional spectrum to one of the axes has a unique interpretation. For gamma–gamma coincidence measurements, the spectrum obtained from this projection yields the gamma-ray spectrum in one detector that corresponds to a specific total energy deposition in both detectors. The total energy deposited may be equal to the Q-value of the reaction studied or the sum of two individual gamma-ray energies, or other energy values of significant meaning. A similar approach was investigated by Eland et al. [4] to study the energy levels of ions.

The diagonal area is mainly defined by two diagonal lines (the window). The slope of the diagonal lines depends on the calibrations of the two detectors. Two factors play a role in determining the window position and width. The first factor is the energy of interest. The energy can either be a single energy or a range of energies. A common energy of interest is the Q-value (total energy) of the reaction studied. If one of the decay schemes of the element of interest has only two gamma rays, we expect that these two gammas will show very clearly when making the diagonal projection.

The second factor that plays a role in determining the window width is the detector resolution. In general, the window should have a constant width along the diagonal. The reason for this constant window width is that, for diagonal summing, the low energy in one detector corresponds to a high energy in the other detector and vice versa. This width will thus be determined by the detector having the worst resolution at the highest energy of interest.

3. Instrumentation and software

The coincidence system used in the present work has been described in detail in an earlier paper [5]. Part of the system is based on Nuclear Instrument Module (NIM) instrumentation standards, while the other part is based on Computer Automated Measurement and Control (CAMAC) instrumentation standards. The computer software KmaxNT [6] is used to manage the CAMAC modules and analyze the acquired data. Through KmaxNT one can process and plot the coincidence and single detector data at the same time.

Two-dimensional data (and plots) are also obtained through KmaxNT.

4. Results

Three examples are presented for using the two-dimensional diagonal summing. The first example is for sulfur. The two detectors used were NaI with dimensions $5'' \times 5''$ and $6'' \times 6''$, placed side by side. A box containing sulfur was placed over the detectors, above which a Cf-252 neutron source was placed in lead shielding. The resolution for these detectors was almost the same, thus a constant width window was used for the diagonal summing. Fig. 1 shows the two-dimensional plot (flat view) of the counts in both NaI detectors. Two windows are outlined, windows 1 and 2. The first window corresponds to the Q-value of the $^{32}S(n,\gamma)^{33}S$ reaction, while the second window corresponds to an energy of 7.800 MeV.

Before presenting the results of the diagonal summing, the total coincidence spectrum in one of the detectors is shown. This can be obtained by performing the first type of projection (summing) stated earlier. Fig. 2 shows the result of projecting

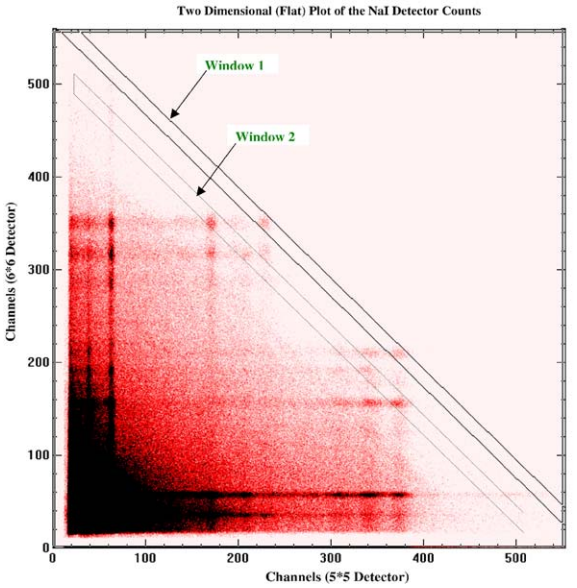

Fig. 1. Two-dimensional plot (flat view) of the counts in both NaI detectors for a sulfur sample.

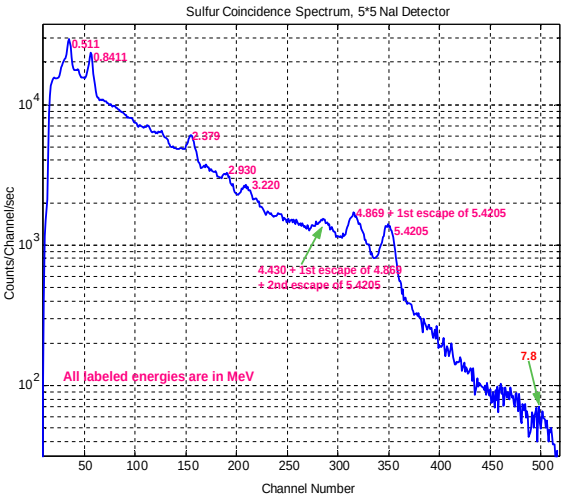

Fig. 2. Two-dimensional full spectrum projection for sulfur.

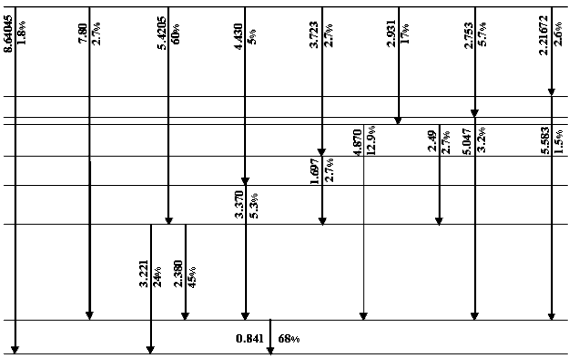

Fig. 3. ^{33}S gamma ray decay scheme (Q-value = 8.641 MeV).

the whole spectrum to the x-axis, i.e. the total coincidence spectrum in the $5'' \times 5''$ NaI detector in coincidence with all events in the $6'' \times 6''$ NaI detector.

The level scheme that is used to analyze this data [7] is shown in Fig. 3, along with the gamma intensities and energies (in MeV). This scheme accounts for approximately 93% of the transitions in the ^{33}S decay.

Next, we observe the resulting spectrum from performing a diagonal summing. Fig. 4 shows the projection of "window 1" and "window 2" in Fig. 1, respectively. The energy of "window 1" corresponds to the Q-value of the $^{32}S(n,\gamma)^{33}S$, 8.641 MeV, while that of "window 2" corresponds

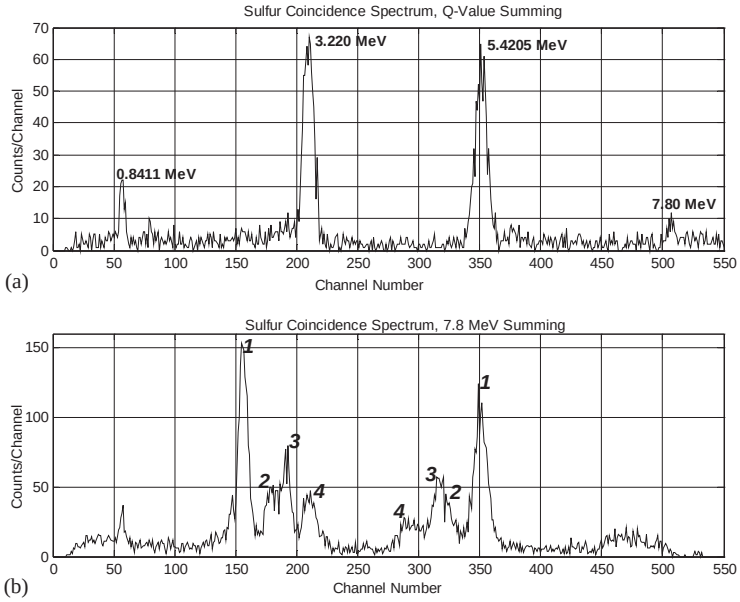

Fig. 4. Q-value and 7.800 MeV diagonal summing for sulfur.

to 7.800 MeV. From Fig. 3, only two schemes contain two gamma rays whose energies add up to the Q-value, 8.641 MeV. The energies of the schemes are 7.800 and 0.841 MeV, and 5.420 and 3.221 MeV. Thus we expect these four gamma rays to show up clearly when performing Q-value diagonal summing. Fig. 4(a) confirms this fact. It also shows that the 5.420–3.221 MeV pair are much more intense than the 7.800–0.841 MeV pair. This is attributed to the difference in scheme probability. Also from Fig. 3, one finds several energies that add up to 7.800 MeV. These energies are labeled in Fig. 4(b) as pair numbers 1–4. The energies corresponding to these pairs are listed in Table 1.

An early paper by Hoogenboom [8] demonstrated a hardware approach to obtain spectra that result from a specific sum of gamma ray energies. This approach was shown to greatly improve the peak resolutions over those obtained from a normal singles spectrum. The diagonal summing approach used here enables us to get the same type of spectrum but with much more accuracy and flexibility. Moreover, to examine another Q-value or sum one can simply interrogate the same data with the different diagonal sum by use of a single set of data from the current electronic system (and KmaxNT). In the hardware approach one would have to perform the experiment again to obtain a different diagonal sum. Fig. 5 shows the Q-value diagonal summing spectrum compared with the total coincidence spectrum. The peak resolution improvement is clearly shown.

Table 1
Energies adding up to 7.800 MeV in the ^{33}S decay scheme

Pair #	Energies in coincidence (MeV)
1	5.420 and 2.379
2	5.047 and 2.753
3	4.870 and 2.931
4	5.420 Compton and 3.221

The second example presented for diagonal summing is for mercury. The NCSU PULSTAR thermal neutron beam was utilized in this experiment. The two NaI detectors ($5'' \times 5''$ and $6'' \times 6''$) were arranged to be facing each other with the sample in between. The sample to detector distance was 20 cm. The sample was elemental natural mercury (12 g m).

Fig. 6 shows the two dimensional plot (flat view) of the counts in both NaI detectors. The outlined

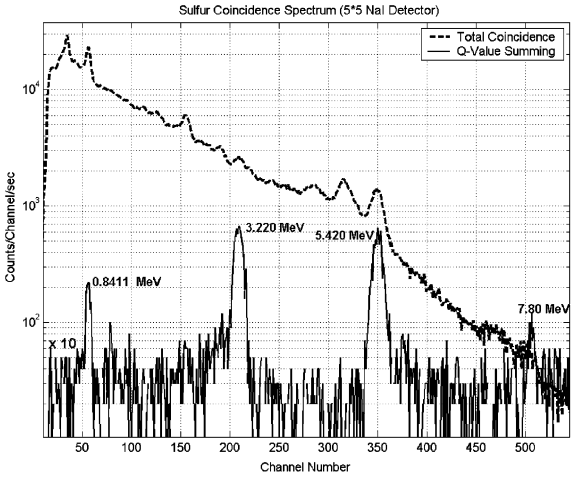

Fig. 5. Comparison of total and Q-value diagonal summing for sulfur.

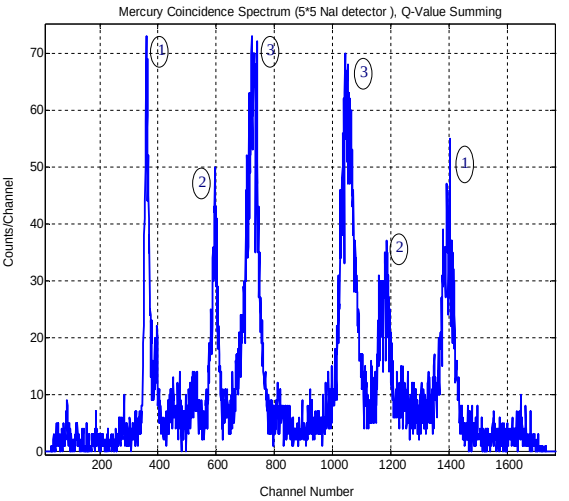

Fig. 7. Q-value diagonal summing for mercury.

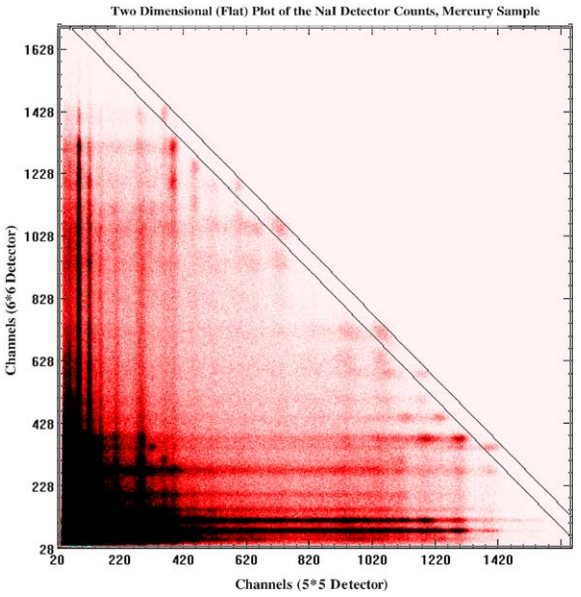

Fig. 6. Two-dimensional plot (flat view) of the counts in both NaI detectors, mercury sample.

diagonal window corresponds to the Q-value (8.028 MeV) of the ^{199}Hg(n,γ)^{200}Hg reaction, which is the dominant (n, γ) interaction with natural mercury. Fig. 7 shows the Q-value diagonal summing for natural Mercury. The dominant coincidence pairs are labeled in the figure and their corresponding energies are listed in

Table 2
Energies adding up to 8.028 MeV in the ^{200}Hg decay scheme

Pair #	Energies in coincidence (MeV)
1	1.571 and 6.457
2	2.639 and 5.387
3	3.288 and 4.739; 3.185 and 4.841

Table 2. A comparison between the Q-value diagonal summing spectrum and the total coincidence spectrum for mercury is shown in Fig. 8. The peak resolution improvement is clearly shown, as is the removal of escape and other peaks that do not sum to give the Q-value.

The third example presents a practical application of two-dimensional diagonal summing. The experimental setup is similar to that in the first example, except that a coal sample was used instead of the sulfur one. The diagonal window energy was set around 8.64 MeV. Upon performing the diagonal summing projection, we found four peaks appearing in the spectrum. Fig. 9 shows the diagonal summing spectrum compared with the total coincidence spectrum. After analyzing the spectrum, we found that the two outer peaks in the diagonal summing spectrum correspond to S-33, identical to those in Fig. 4(b), and the two inner peaks correspond to Si-28 which has a Q-value of

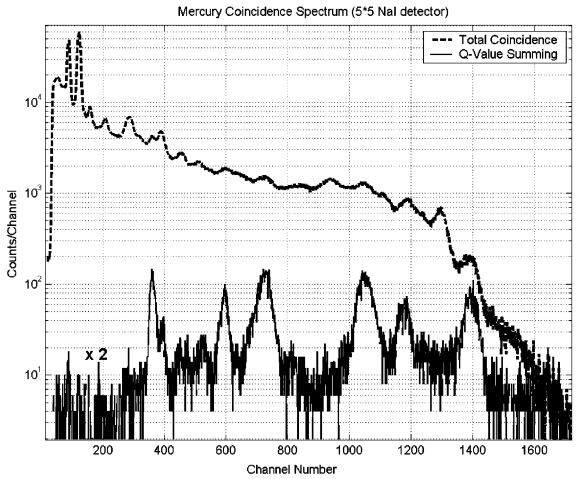

Fig. 8. Comparison of full and diagonal summing for mercury.

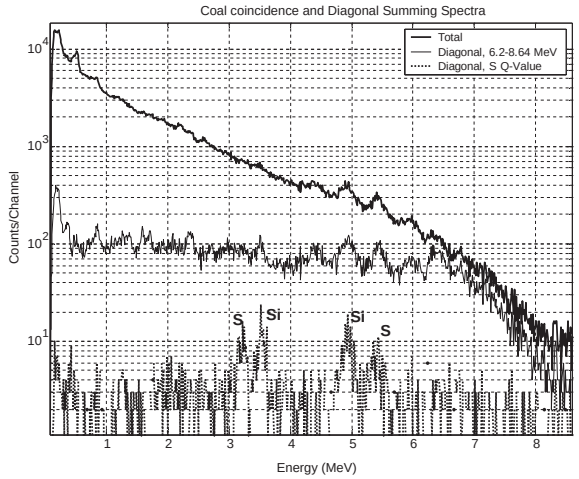

Fig. 9. Comparison of full and diagonal summing spectra for coal.

8.473 MeV. The other diagonal summing spectrum shown in Fig. 9 corresponds to an energy window from 6.200 to 8.700 MeV. It is clear from this spectrum that summing over a range of high energies eliminates the coincidences of interfering low-energy gamma-ray pairs, thus increasing signal-to-noise (S/N) ratio.

The diagonal summing spectra presented in Fig. 9 are good examples of where the Library Least Squares (LLS) approach can be applied [5]. The LLS approach is very useful for inverse spectral problems such as determining elemental amounts from gamma-ray, X-ray or prompt gamma-ray spectra. The diagonal summing spectra from either a narrow or a wide window, such as those in Fig. 9, may be used with the LLS approach. The optimum diagonal window width will depend on the specific application and/or the element of interest. For example, if sulfur is of interest the optimum window may be that corresponding to the energy range from 7.800 to 8.641 MeV. In addition to the low-energy Compton scattering contribution within this window, it contains most of the high-intensity sulfur gamma rays. The authors are investigating this approach.

5. Conclusions

The concept of two-dimensional diagonal summing has been introduced and demonstrated for the two elements sulfur and mercury. The main advantages demonstrated for this approach are:

1. improved peak resolution and
2. very low background (Compton continuum).

In addition to these advantages, it is shown that different diagonal sums can be examined by use of the same set of data with the current electronic system (and software).

Possible uses of the two-dimensional diagonal summing are identifying coincidence schemes, performing elemental analysis, and identifying trace elements in bulk samples. The results obtained indicate that the approach is very promising for these purposes.

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