High-resolution boosted reconstruction of γ-ray spectra postprint

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Abstract

The Direct Demodulation Method (DDM) was applied to reconstruct γ-ray spectra. A boosted Richardson-Lucy iteration algorithm was introduced into the DDM framework. The Monte Carlo method (specifically GEANT4) was employed to calibrate the response function and establish the response matrix. Initially, the Gaussian function was used to model the total energy peak. Spectral lines were simulated using nine Gaussian functions. Subsequently, DDM was applied to reconstruct the simulated spectral lines and determine peak positions and areas. Compared with the original spectra, for cases where the peak position interval was approximately 1/3 of the full width at half maximum (FWHM), the error in the reconstructed peak position was 2 channels. The remaining peaks could be accurately identified. The relative errors for all peak areas were less than 4%. Subsequently, three key factors—including noise, background, and response matrix—were investigated. Finally, DDM was applied to calibrate a field NaI gamma spectrometer. The errors for U, Th, and K were less than 5%. Comprehensive studies have demonstrated that reconstructing gamma-ray spectra using DDM is feasible. DDM can significantly improve the effective energy resolution of gamma spectrometers, effectively decompose doublets with peak intervals of 1/3 FWHM, and accurately identify peaks and calculate their areas. DDM exhibits strong noise suppression capabilities but is significantly influenced by background. Furthermore, DDM can enhance the accuracy of qualitative and quantitative analysis when combined with conventional spectrum analysis methods.

Full Text

Preamble

High-Resolution Boosted Reconstruction of γ-Ray Spectra

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Abstract: The Direct Demodulation Method (DDM) was applied to reconstruct γ-ray spectra, with a boosted Richardson-Lucy iteration introduced into the DDM framework. Monte Carlo simulation using GEANT4 was employed to calibrate the response function and establish the response matrix. First, a Gaussian function was used to model the total energy peak, and a spectral line was simulated using nine Gaussian functions. DDM was then applied to reconstruct this simulated spectral line and determine peak positions and areas. Compared with the original spectrum, for cases where the peak position interval was about 1/3 full width at half maximum (FWHM), the error in reconstructed peak position was 2 channels, while all other peaks could be searched accurately. The relative errors of all peak areas were less than 4%. Subsequently, three key factors—noise, background, and response matrix—were discussed. Finally, DDM was applied to calibrate a field NaI gamma spectrometer, yielding errors of less than 5% for U, Th, and K. Comprehensive studies demonstrate that reconstructing γ-ray spectra with DDM is feasible. DDM can significantly pseudo-improve the energy resolution of gamma spectrometers, effectively decompose doublets with peak interval as small as 1/3 FWHM, and accurately search peaks and calculate areas. DDM exhibits strong noise suppression capability but is greatly influenced by background. When combined with conventional spectrum analysis methods, DDM can improve the accuracy of both qualitative and quantitative analysis.

Keywords: Direct Demodulation Method (DDM), Monte Carlo, GEANT4, Reconstruction, Doublets
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Introduction

Gamma energy spectrum analysis is a technique that uses a gamma spectrometer to measure γ-rays from radionuclides and determines the categories and contents of particular radionuclides by processing the measured γ-ray spectra. Data processing generally includes smoothing, background deduction, peak searching, calculation of peak areas, and energy calibration. Peak position and peak area are the keys to qualitative and quantitative analysis, respectively. Traditional

peak searching methods [1] include the IF function method, Gaussian product function method, derivative method, covariance method, and symmetrical zero area transformation method. Although these methods can achieve high precision for strong radioactive γ-ray spectra and singlets, they perform poorly for weak radioactive γ-ray spectra and doublets. For weak radioactive γ-ray spectra in particular, the IF function method, Gaussian product function method, and covariance method are not desirable [2], while derivative and symmetric zero area methods have limited effectiveness.

Most traditional peak area methods—such as the total peak area (TPA) method, Covell peak area method, Wasson peak area method, Sterlinski peak area method, Wasson-Sterlinski peak area method, Quittner peak area method, W-S-Q peak area method, and Q-S peak area method—require determination of peak position and boundaries and can only be applied to singlets [3]. The curve fitting method performs better [4, 5]. Its basic principle involves establishing a mathematical model or function to describe the shape of the total energy peak and baseline, determining the function parameters from experimental data, and obtaining the peak area by integrating the function. However, due to the complexity of detectors and detection environments, measured γ-ray spectra are often too complicated for reliable function determination, which severely limits the application of curve fitting methods for peak area calculation.

In 1997, de-convolution was proposed for processing γ-ray spectra [6]. For ill-posed problems, conventional de-convolution methods such as the maximum entropy method [7] are sensitive to input data errors, where small errors can cause large oscillations. Fortunately, Tikhonov et al. studied ill-posed problems in 1977 on a strict mathematical basis by introducing regularization theory and methods [8]. The idea is to transform ill-conditioned problems into well-posed problems to ensure acceptable and stable solutions. Regularization generally falls into three categories: least squares, smoothing methods, and iterative approaches. The least-squares method is vulnerable to premature convergence and yields solutions with large errors [9]. The degree of smoothing is difficult to control, and smoothing itself introduces errors [10]. In iterative approaches, successive approximations are generated to converge to an appropriate solution. Perhaps the most significant feature of iterative methods for unfolding applications lies in their ability to readily incorporate most major physical implications of the problem description while avoiding explicit calculation of the inverse matrix [11]. This is important because most response matrices are ill-conditioned, and errors in inverse matrix elements can be prohibitive.

Three commonly used regularization (iteration) de-convolution methods are Gold [12], Richardson-Lucy [13], and maximum posteriori algorithms [14]. The Direct Demodulation Method (DDM) [15, 16] was proposed by Li Tibei and Wu Mei. Its basic principle uses known physical conditions to control the iterative process for solving the modulation equation. The Richardson-Lucy iteration converges to the maximum likelihood solution. Compared with conventional de-convolution methods, DDM is rarely restricted by the shape of γ-ray spectra,

radioactive intensity, or input error, as it takes full advantage of known information and utilizes nonlinear physical constraint conditions. DDM has succeeded in high-energy astronomy for reconstructing astrophysical images with low signal-to-noise ratio (SNR), low statistics, and low resolution [17–19]. However, there are few reports on DDM applications for γ-ray spectra. In this paper, we propose utilizing DDM to reconstruct γ-ray spectra, with Monte Carlo simulation performed to calibrate response functions and establish the response matrix. The purpose of this study is to improve the energy resolution and accuracy of γ-ray spectra analysis.

II. The Direct Demodulation Method

The input and output of a gamma spectrometer can be regarded as a complete system. Incident radiation can be imaged as a sum of δ-functions, so at the system output the spectrum represents a linear combination of δ-functions with various amplitudes located at various channel functions, which are blurred by the system response functions. The purpose of DDM is to eliminate the influence of the response function to the utmost extent, ideally obtaining a complete γ-ray spectrum consisting of δ-functions.

Let us suppose the intensity distribution function of the source is f(x). From a signal processing perspective, the measurement process of a spectrum can be regarded as the modulation process of the spectrum from the radioactive source. The measured γ-ray y(j) is the modulated result. The modulation equation can be expressed as:

$$\int h(j, x)f(x)dx = y(j),$$

where h(j, x) represents the system response function. The matrix equation can be expressed as:

$$HF = Y,$$

where H is the system response matrix. For γ-ray spectra, H is a lower triangular Toeplitz matrix:

[H matrix representation]

Equation (2) means that the spectrum of the radioactive source F is modulated by the gamma spectrometer, and the measured spectrum Y is obtained. The reconstruction process is the inverse process: demodulating the spectrum Y to reconstruct the spectrum of the radioactive source F.

For gamma spectrometry, y(j) and f(x) are both nonnegative discrete points, which is vitally important for DDM. Additionally, h(j, x) is discretized in numerical calculation. Hence, the discrete form of Eq. (1) can be expressed as:

$$\sum h(k, j)f(j) = y(k),$$

where h(k, j) is the element at the kth row and jth column of response matrix H, f(j) is the jth element of F, and y(k) is the kth element of Y. However, Eq.

(4) is an ill-posed problem, so DDM is applied to solve it.

The Richardson-Lucy iteration algorithm (R-L) is based on Bayesian statistical theory and can be expressed as:

$$f\hat{}(n+1)(j) = f\hat{}(n)(j) \cdot [h(k, j)d\hat{}(n)(k)/ h(i, j)],$$

where $d\hat{}(n)(k) = h(k, j)f\hat{}(n)(j)$. However, Eq. (5) is prone to "premature" convergence, so an enhanced Richardson-Lucy iterative algorithm [8] is applied in this paper.

The nonlinear physical constraints, including upper and lower limits, can be expressed as:

$$f\hat{}(n)(j) > u(j) \quad f\hat{}(n)(j) = u(j);$$
$$f\hat{}(n)(j) < b(j) \quad f\hat{}(n)(j) = b(j).$$

Obviously, each element of F is less than the upper limit, so only the lower limit needs to be considered. For γ-ray spectra, background is regarded as the lower limit b(j).

Equations (5) and (6) constitute a complete representation of DDM. Referring to Ref. [8], an exponential factor p was introduced into DDM. The principles of enhanced DDM can be expressed as follows:

Step 1: For n = 0, set the iterative initial value $f\hat{}(0) = [1, 1, \cdots, 1]\hat{}T$.
Step 2: Set the number of iterations L and cycle number R.
Step 3: Initialize the cycle number r = 1.
Step 4: According to Eq. (4), calculate and seek the solution $f\hat{}(L)$.
Step 5: Introduce parameter p, and $f\hat{}(0)(i) = [f\hat{}(L)(i)]\hat{}p$ where p (1, 2), i = 0, 1, ···, N-1.
Step 6: Introduce physical constraints.
Step 7: If r = R, then stop calculating. Otherwise, r = r + 1 and continue to Step 4.

III. Results and Evaluation

The difficulty evaluation for separation of doublets from the IEC standard [21] is given in Table 1 . Referring to this standard, Gaussian functions were used to simulate nine full-energy peaks, with their positions, heights, and areas listed in Table 2 . The synthetic spectrum is shown in Fig. 1 Figure 1: see original paper. From Table 2, the largest height ratio is 10:1, and the minimum interval between peak positions is less than 1/3 FWHM. All cases except the area ratio of 1:100 in Table 1 are considered. The spectral line was reconstructed by applying DDM with 1000 (L = 50, R = 20, p = 1.8), 5000 (L = 100, R = 50, p = 1.8), 10000 (L = 200, R = 50, p = 1.8), and 50000 (L = 500, R = 100, p = 1.8) iterations, respectively. Peak areas were then calculated by accumulating counts for "isolated" singlets in the reconstructed spectrum. The result after 50000 iterations is shown in Fig. 1(b).

The width of the reconstructed peak decreased with increasing number of iterations. Theoretically, a complete spectrum consisting of δ-functions (single pulses) can be obtained after a large number of iterations, with corresponding pulse counts representing peak areas. However, more iterations consume more time and memory. Thus, from an efficiency perspective, iteration should be stopped when doublets are completely separated. In this case, peak areas are not the corresponding pulse counts but the sum of counts for a few "isolated" points. The results demonstrate that DDM can effectively decompose peaks overlapped by as little as 1/3 FWHM, which represents an extremely difficult task.

Table 3 lists the peak positions and areas of the DDM-reconstructed spectrum. For an area ratio of 1:1 and peak position interval of about 1/3 FWHM, the error in peak position is 2 channels, while it is 1 channel for an area ratio of 1:10 and peak position interval of about FWHM. The peak area error is less than 4% for all cases, indicating that positions and areas can be precisely obtained with DDM.

IV. Discussion

A measured γ-ray spectrum typically contains noise and background, making it necessary to study their influence on DDM.

A. Noise

A Gaussian peak was added with random noise (Fig. 2(a)) and Gaussian noise (Fig. 2(c)) at amplitudes of 3% and 1% of the Gaussian peak height, respectively. The results are shown in Figs. 2(b) and 2(d), respectively. DDM exhibits a strong inhibitory effect on noise because least-squares, smoothing methods, and iterative approaches can solve ill-conditioned problems, and the Richardson-Lucy iteration algorithm intangibly suppresses noise during the iteration process. If noise is not severe, a spectrum can be reconstructed without denoising.

B. Background

A spectrum was added with straight-line, oblique-line, and step backgrounds, respectively. The DDM-reconstructed results are shown in Fig. 3 [Figure 3: see original paper]. The spectra were not reconstructed correctly, indicating that background affects the accuracy of reconstructed spectra. However, research shows that if the response matrix can be obtained under the same background conditions, or if the background can be removed from both the measured spectrum and response matrix using the same method, the spectral lines can be reconstructed accurately.

C. Establishment of Response Matrix with GEANT4

Experiments have shown that the response function of a spectrometer system is strongly dependent on the energy of incident γ-rays. While no unanimous theories or empirical expressions exist, the response function can be obtained experimentally. Ideally, the response function should be calibrated using standard radioisotope sources with strong single peaks distributed uniformly across the entire energy range. However, obtaining so many standard sources is impractical, and γ-ray peaks cannot be distributed so uniformly. Fortunately, the Monte Carlo method can correctly simulate the response function of γ-ray spectra [22, 23], allowing artificial control of source distribution and energy.

GEANT4 [24] can be used for neutron, photon, electron, or coupled neutron/photon/electron transport, as well as protons, and is capable of calculating eigenvalues for critical systems. For photons, the code accounts for incoherent and coherent scattering, fluorescent emission after photoelectric absorption, absorption in pair production with local emission of annihilation radiation, and bremsstrahlung. The photon energy regime extends from 1 eV to 100 GeV for Rayleigh and Compton effects, down to the lowest binding energy for each element for photoelectric and ionization processes, and down to 10 eV for bremsstrahlung. GEANT4 was employed in this work.

To coincide with experiments, detector properties must be revised before simulation. ^{241}Am (0.06 MeV), ^{57}Co (0.122 MeV), ^{22}Na (0.51 MeV), ^{137}Cs (0.661 MeV), and ^{24}Na (1.38 MeV, 2.75 MeV) were selected. Crystal dimensions were adjusted to ensure that the spike and full-energy peak efficiency of the measured spectrum coincided with simulation results. Then, based on energy calibration data, the corresponding energy per channel was chosen to simulate the response function, which was adjusted according to the multi-channel analyzer. Analog data for each single peak were normalized as the response function for that energy. The response function needs to be converted to a response matrix for DDM use. Since the measurement spectrum is the convolution of input data and response function, the response function is reflected, zero-filled, and gradually shifted to obtain a series of two-dimensional vectors that constitute the response matrix.

V. Calibration of NaI Gamma Spectrometer Based on DDM

DDM was used to calibrate a field NaI gamma spectrometer. Generally, spectrum stripping is employed, with calibration principles described in Ref. [25]. An IED-3000A NaI gamma spectrometer (No. 104) was used, featuring a NaI crystal with dimensions of 75 mm $\times$ 75 mm and an energy resolution of 10.3% at 661 keV. The measuring time was 3600 s. Substance contents of the quasi-saturation model are listed in Table 4 , as measured by the Analysis and Test Center of China Geological Survey.

The measured spectrum of the hybrid model is shown in Fig. 4 Figure 4: see original paper. SNIP [26] was applied to deduct background, and DDM was used to reconstruct the spectrum. The result after 50000 iterations is shown in Fig. 4(b). Calibration coefficients and net peak areas for 1.46 MeV (^{40}K), 1.76 MeV (^{214}Bi of U series), and 2.62 MeV (^{208}Tl of Th series) are listed in Table 4.

Table 5 shows elemental analysis results for a γ-ray analysis standard obtained using DDM and the spectrum stripping method (SSM). The DDM results show smaller deviations from the given values for U, Th, and K, being -1.02%, -4.19%, and -2.41%, respectively, while the SSM results deviated by -8.06%, 10.44%, and 8.84%, respectively.

VI. Conclusion

Due to its response function and iterative algorithm, DDM is rarely restricted by the shape of γ-ray spectra or radioactive intensity. DDM can significantly pseudo-improve energy resolution, precisely search peaks, calculate areas, and improve gamma spectrometer performance without hardware cost. It is worth emphasizing that the response matrix is the key to DDM. However, in practice, it is tedious and difficult to use standard radioactive sources to establish the response matrix. Therefore, GEANT4 was proposed to calibrate the response function, making it convenient, safe, and effective to employ the Monte Carlo method for response matrix establishment.

Theoretically, DDM can pseudo-improve energy resolution to infinity after a large number of iterations, but larger iterations consume more time and memory. Thus, from an efficiency perspective, iteration should be stopped when doublets are completely separated. It should be emphasized that DDM is theoretically appropriate not only for γ-ray spectra but for any other spectrum as long as the response matrix can be correctly established. In fact, DDM can be applied not just to NaI(Tl) detectors but to any other detector. It is time-consuming to establish the response matrix and run DDM, especially for complex spectra, so DDM is typically used for offline analysis. Future research will focus on improving running speed.

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