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I. N. Izosimov *

APPLICATION OF THE METHOD OF MOMENTS FOR THE DOUBLET AND TRIPLET ANALYSIS IN THE RADIATION SPECTRA

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* E-mail: izosimov@jinr.ru

Изосимов И. Н. Е6-2023-29

Применение метода моментов для анализа дублетов
и триплетов в спектрах излучений

При обработке спектров обычно достаточно определить только положение пиков и их площади. Чтобы использовать метод моментов для анализа дублетов, необходимо сначала построить (откалибровать) зависимость второго и третьего моментов для одиночных пиков от энергии соответствующего излучения. После такой калибровки перекрывающиеся пики могут быть разделены. Чтобы разделить пики в дублете с помощью метода моментов, необходимо решить квадратное уравнение. Чтобы разделить пики в триплете, необходимо иметь соответствующие калибровочные кривые уже до пятого момента включительно и решить кубическое уравнение. Представлены примеры разделения перекрывающихся пиков при анализе спектров гамма-излучения.

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Izosimov I. N. E6-2023-29

Application of the Method of Moments for the Doublet
and Triplet Analysis in the Radiation Spectra

When processing spectra, it is usually sufficient to determine only the positions of the peaks and their areas. To use the method of moments for doublet peaks analysis, it is necessary to first construct (calibrate) the dependence of the second and third moments for single peaks on the energy of the corresponding radiation. After such calibration overlapping peaks can be separated. To separate peaks in doublet using the method of moments, it is necessary to solve a quadratic equation. To separate peaks in triplet, it is necessary to have appropriate calibration curves already up to the fifth moment inclusive and solve a cubic equation. Examples of the separation of overlapping peaks in the analysis of gamma-ray spectra are presented.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

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INTRODUCTION

In the currently used programs for spectra analysis, the shape of peaks is described either by a model function or using a table. The deviation of the experimental peak form from the model one leads to the appearance of systematic errors in the analysis of overlapping peaks. When tabulating the shape, it is difficult to take into account the dependence of the peak shape on energy. For spectra analysis, it is usually sufficient to determine only the positions of the peaks and their areas, but when using parametric methods, it is necessary to determine the full set of parameters describing the shape of the peak. Therefore, methods that do not use an explicit description of the shape of peaks are interesting. Such methods are called distribution-free or nonparametric.

The moments of distribution carry sufficiently complete information about the shape of the instrument lines and they can be used both for the analytical description of the shape of the peaks when using the minimum χ^2-method, and for the direct separation of overlapping peaks by solving simple algebraic equations without using any model assumptions about their shape [1]. If the dependence of the moments on the energy obtained after determination of the moments for single peaks is known, overlapping peaks can be separated. To separate doublets using the method of moments, it is necessary to solve a quadratic equation; to separate triplets, a cubic equation. To use the method of moments for peak separation in doublet, it is necessary to first construct calibration curves of the dependence of the second and third moments for single peaks on the energy of the corresponding radiation. To separate peaks in triplet it is necessary to have appropriate calibration curves already up to and including the fifth moment. The report provides examples of the separation of overlapping peaks in the analysis of gamma-ray spectra. Methods for estimating the errors of the results and the applicability of the method of moments for separating overlapping peaks are discussed.

1. SAMPLE MOMENTS AND PEAK SHAPE IN THE RADIATION SPECTRA AND CALIBRATION CURVES FOR THE SECOND AND THIRD MOMENTS

The position of the peak is usually associated with the position of the maximum of the model function $f(i)$, but since it is necessary to find estimates that do not use parametric function for the peak shape (Fig. 1) in the experimental spectrum $S(i)$, it is necessary to use another definition of

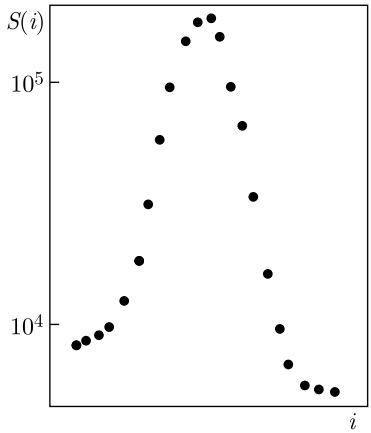

Fig. 1. The shape of the peak (background included) in the γ-ray spectrum

this value. For the spectra we are considering, $S(i)$ as a discrete random variable has a Poisson distribution in i channel. It is natural to associate the mathematical expectation $M\{S(i)\}$ with the energy of the peak, and use the sample average as a statistical estimate of the peak position. Then for the position P and the area B of a single peak (if there is no background) we have

$$\overline{P} = \sum_i S(i) i / \overline{S}, \quad \overline{B} = \sum_i S(i). \tag{1}$$

The line at the top indicates estimates of random variables. The variance of the estimate of the value x will be denoted by $D\{x\}$, and $\sqrt{D\{x\}} - \Delta x$. Then $\Delta B = \sqrt{B}$, since $D\{B\} = \overline{B}$. The position P according to the definition given above is the first sampling moment, hence [2, 3] $\Delta P = M_2/B$, where M_2 is the second central moment of $S(i)$ distribution. For the $S(i)$ distribution, it is possible to estimate the higher central moments M_n and their dispersions $D\{M_n\}$ according to the formulas [2, 3].

The highest moments are associated with the shape of the peak. This relationship can be expressed analytically using decompositions used in probability theory [2–4].

The method of moments is quite suitable for describing the shape of an instrument line in different spectra. If the dependence of the moments on the energy obtained after calculating the moments for single peaks is known, the overlapping peaks can be separated by changing only the positions and areas of the components. When using moments to account for the final channel width, moments of the density $S(i)$ grouped by channels M'_n and the moments

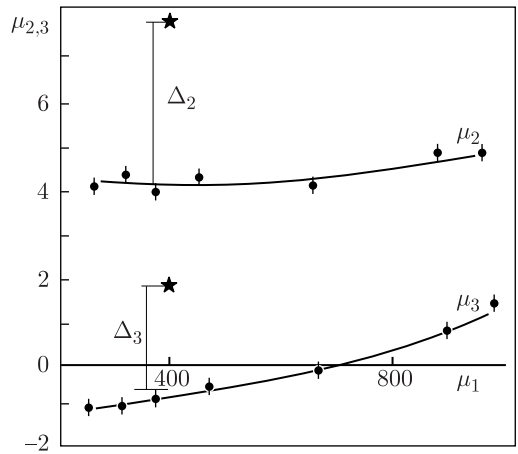

Fig. 2. The dependence of the moments μ_2 and μ_3 on the μ_1 for single peaks and $\Delta_i = M_i - \mu_i$, where M_i is the measured moment for doublet [1]. The value of μ_1 is related to the γ-ray energy by using a calibration procedure [5]

of the initial density M_n are connected by simple relations [2–4]. For the second moments one has

$$M_2' = M_2 + 1/12. \tag{2}$$

Figure 2 shows an example of the dependence of the central moments on the energy for the γ-ray spectrum measured using a semi-conductor HPGe detector. It can be seen from Fig. 2 that the moments μ_2 and μ_3 change quite slowly with increasing the γ-ray energy. The points marked with stars correspond to a section of the spectrum containing two overlapping peaks. Obviously, the deviations Δ_2 and Δ_3 are related to the areas and positions of these peaks, and using this relationship, it is possible to separate overlapping peaks without minimizing the χ^2 function [1].

2. DETERMINATION OF THE POSITION AND AREAS OF PEAKS FOR DOUBLET

The measured or instrumental spectrum is the distribution density $\rho(V)$ of the amplitudes of the pulses V, which are caused by the measured radiation in the detector. The true spectrum or density of the probability distribution of the detected radiation with energy E is determined from the calibration of the detector with radiation energy, i.e., the determination of the instrumental response function (response function) [1].

Consider the case of two overlapping peaks. Let's assume for the doublet $M_1 = 0$; that is, the positions of the doublet components will be counted from the center of gravity of the multiplet. In this case, the measured spectrum,

assuming that the shape of the peaks in the multiplet is the same, can be written as follows:

$$\rho(V) = a_1\rho_1(V - b_1) + a_2\rho_1(V - b_2). \tag{3}$$

We determine the values $\Delta_i = M_i - \mu_i$ for the analyzed doublet (Fig. 2), where M_i is the corresponding measured moment for the doublet, μ_i is the moment known from calibration experiments for single peaks. We introduce symmetric polynomials $\sigma_1 = b_1 + b_2$, $\sigma_2 = b_1 b_2$. Using the expressions and the definition of moments for the doublet, we obtain the following equations for determining the values of b_1, b_2, a_1, a_2:

$$\Delta_2 = \sigma_2, \quad \Delta_3 = \sigma_1\sigma_2, \tag{4}$$

solving which we separate doublet peaks:

$$\begin{aligned} b_{1,2} &= (1/2)\left(\Delta_3/\Delta_2 \mp \sqrt{\Delta_3^2/\Delta_2^2 + 4\Delta_2}\right), \\ a_1 &= b_2 \Big/ \sqrt{\Delta_3^2/\Delta_2^2 + 4\Delta_2}\ , \quad a_2 = 1 - a_1. \end{aligned} \tag{5}$$

Note that to separate the peaks of the doublet, it is necessary to have a calibration curve (Fig. 2) up to and including the third moment.

Knowing the a_k and a_k, we can determine the areas B_k and positions P_k of the doublet components:

$$B_k = M_0 a_k, \quad P_k = M_1 + b_k. \tag{6}$$

To determine the errors, neglecting correlations, we introduce the β_i values and differentiate (5). After that, using standard algorithms [2–4], we get

$$\begin{aligned} \beta_2 &= (2 - (\Delta_3^2/\Delta_2^2))/\sqrt{\sigma_1^2 - 4\sigma_2} - \Delta_3/\Delta_2, \\ \beta_3 &= (1/\Delta_2) + \Delta_3/(\Delta_2^2\sqrt{\sigma_1^2 - 4\sigma_2}\,), \end{aligned} \tag{7}$$

$$D\{b_1\} = (1/2)[\beta_2^2(D\{M_2\} + D\{\mu_2\}) + \beta_3^2(D\{M_3\} + D\{\mu_3\})], \tag{8}$$

$$D\{b_2\} = D\{b_1\}(a_2/a_1),$$

$$D\{B_k\} = D\{B_0\}B_0/B_k. \tag{9}$$

The table and Fig. 3 show examples of the separation of two overlapping peaks according to the above formulas. Doublet was obtained by superposition of the two single lines. The shape of the line is shown in Fig. 1. After

Example of analysis of two overlapping peaks

Area of peak		Position of peak $\times 10^3$	
Set	Determined	Set	Determined
485154	490700 ± 4900	201157	201141 ± 17
45274	41300 ± 4100	204125	203810 ± 200

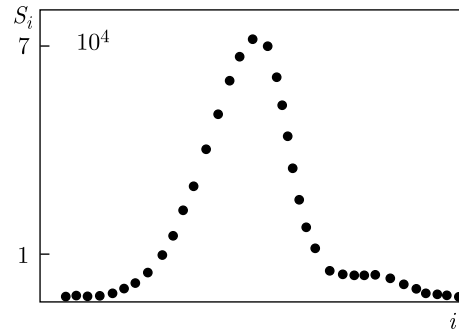

Fig. 3. Doublets obtained by overlapping two single peaks without background

superposition the areas and position of single lines for doublet were determined by formulas (5).

3. DETERMINATION OF THE POSITION AND AREAS OF PEAKS FOR TRIPLET

The measured spectrum for triplet, assuming that the shape of the peaks in the multiplet is the same, can be written as follows:

$$\rho(V) = a_1\rho_1(V - b_1) + a_2\rho_1(V - b_2) + a_3\rho_1(V - b_3). \tag{10}$$

Using the expressions for calculating the moments of the triplet M_i and the values known from the calibration curve for single peaks μ_i, we obtain the following relations:

$$\sum_{i=1}^{3} a_i = 1, \quad \sum_{i=1}^{3} a_i b_i = 0, \quad \sum_{i=1}^{3} a_i b_i^2 = \Delta_2, \quad \sum_{i=1}^{3} a_i b_i^3 = \Delta_3, \tag{11}$$

$$\sum_{i=1}^{3} a_i b_i^4 = \Delta_4 - 6\mu_i\Delta_2 \equiv L_4, \quad \sum_{i=1}^{3} a_i b_i^5 = \Delta_5 - 10(\mu_3\Delta_2 + \mu_2\Delta_3) \equiv L_5. \tag{12}$$

Introducing the corresponding symmetric polynomials:

$$\sigma_1 = b_1 + b_2 b_3, \quad \sigma_2 = b_1 b_2 + b_2 b_3, \quad \sigma_3 = b_1 b_2 b_3, \tag{13}$$

and substituting (13) into (11) and (12), we obtain the equations for calculating $\sigma_1, \sigma_2, \sigma_3$:

$$\begin{gathered} \Delta_3 = \sigma_3 + \sigma_1\Delta_2, \quad L_4 = (\sigma_1^2 - \sigma_2^2)\Delta_2 + \sigma_1\sigma_3, \\ L_5 = (\sigma_1^3 - 2\sigma_1\sigma_2 + \sigma_3)\Delta_2 + \sigma_3(\sigma_1^2 - \sigma_2), \end{gathered} \tag{14}$$

solving which we get

$$\sigma_1 = [L_5 + \Delta_2\Delta_3 + (\Delta_3/\Delta_2)L_4]/[L_4 - \Delta_2^2 - (\Delta_3^2/\Delta_2)],$$
$$\sigma_2 = (\sigma_1\Delta_3 - L_4)/\Delta_2, \quad \sigma_3 = \Delta_3 - \Delta_2\sigma_1. \tag{15}$$

By entering the variables $u = b_2 + b_3$ and $v = b_2b_3$, we obtain a cubic equation for determining the value of u:

$$u^3 - 2\sigma_1u^2 + (\sigma_1^2 + \sigma_2)u - \sigma_1\sigma_2 + \sigma_3 = 0, \tag{16}$$

and the corresponding value of v:

$$v = \sigma_2 - \sigma_1u + u^2. \tag{17}$$

Since the original equations are symmetric, it is necessary to find one root of the cubic equation, which can be done numerically. If the solution to Eq. (16) is found, then the desired parameters b_1, b_2, b_3 can be determined:

$$b_1 = \sigma_1 - u, \quad b_{2,3} = (u \pm \sqrt{u^2 - 4v})/2. \tag{18}$$

The a_i parameters are found as solutions of a system of linear equations:

$$a_2 = (\Delta_2 + b_1b_2)/[(b_2 - b_1)(b_3 - b_1)], \quad a_3 = [b_3 + a_2(b_2 - b_3)]/(b_3 - b_1),$$
$$a_1 = 1 - a_3 - a_2. \tag{19}$$

The areas B_k and positions P_k of the triplet components are determined by using formula (6).

It should be noted that in order to analyse a triplet, it is necessary to have appropriate calibration curves (of the type shown in Fig. 2) already up to and including the fifth moment.

The errors of the obtained values can be estimated in a standard way [1–4]. If the contributions of the fourth and fifth moments to the error value can be neglected, then the corresponding formulas for estimating the error are simplified [1]:

$$\Delta P_1 = \Delta P\sqrt{S/3B_1}, \quad \Delta P_2 = \sqrt{B_1/B_2}, \quad \Delta P_3 = \sqrt{B_2/B_3},$$
$$\Delta a_2 = \sqrt{D\{M_2\} + D\{\mu_2\}}\,/[(p_2 - p_1)(p_2 - p_3)],$$
$$\Delta a_1 = \Delta a_2\sqrt{(p_1 - p_3)/(p_3 - p_1)}, \tag{20}$$
$$\Delta a_3 = 0.5[\Delta a_2\sqrt{a_1/a_3} + \Delta a_3\sqrt{a_2/a_3}].$$

DISCUSSION AND CONCLUSIONS

The examples given show that sample moments can be used to separate overlapping peaks by solving simple algebraic equations without using any model assumptions about their shape. To use the method of moments for doublet peaks separation, it is necessary to first construct the special calibration curves for second and third moments. For triplet peaks separation

the calibration curves must be obtained for moments up to the fifth. The necessary information for constructing such curves and for dependence of the sample moments on radiation energy can be obtained from the analysis of single peaks with known energy. Proper standard sources of radiation must be used for calibration procedure [5].

When processing spectra by the method of moments, it is important to ensure the same marking of all intervals (selection of the beginning and end of the multiplets). In cases where increased accuracy of processing is required, the boundaries of the peaks can be adjusted using estimates of the first two or three sample moments obtained after preliminary rough marking, performed, for example, using an automatic peak search program. Errors associated with marking and background should be included in the analysis for variances of estimates of sample moments based on the selected method of marking the spectrum.

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Издательский отдел Объединенного института ядерных исследований
141980, г. Дубна, Московская обл., ул. Жолио-Кюри, 6.
E-mail: publish@jinr.ru
www.jinr.ru/publish/