Least square fitting of low resolution gamma ray spectra with cubic B-spline basis functions*

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Abstract In this paper, the least square fitting method with the cubic B-spline basis functions is derived to reduce the influence of statistical fluctuations in the gamma ray spectra. The derived procedure is simple and automatic. The results show that this method is better than the convolution method with a sufficient reduction of statistical fluctuation.

Key words B-spline basis functions, least square fitting, gamma-ray spectra

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1 Introduction

In general, the gamma ray spectra obtained by NaI detector always contains statistical fluctuations. The spectra can be considered to be a linear sum of two components: $F(x)=S(x)+N(x)$, where $S(x)$ is the spectra information and $N(x)$ is the fluctuations which can be considered as high frequency noise. The objective of analyzing gamma-ray spectra is to extract the useful information $S(x)$ from the measured data while minimizing the influence of fluctuations $N(x)$ which can cause error on both nuclide recognition and quantitative analysis. Up to know, a number of mathematical procedures have been developed for processing detected data, for example, the polynomial fitting[1—4], the Fourier transformation[5—9] and the convolution method[1, 10—17]. Among these methods, the convolution operation, as described by Savitsky and Golay[1], is considered as the most widely used approach in which each data point is in effect replaced by the convolution value of a filter with a small number of adjacent data points.

However, applications of these methods are not always efficacious, especially in the case of intensive noised spectra as Fig. 1. The polynomial fitting method applied to remove the statistical fluctuation always comes along with some problems, for example, spectral distortion, weak peaks easily lost and false peaks generated that can rise calculation error in background determination, peak searching, fitting, etc. The Fast Fourier Transform (FFT) methods have no intrinsic advantage in the application of this area because of little energy information reserved in the transformed data[18].

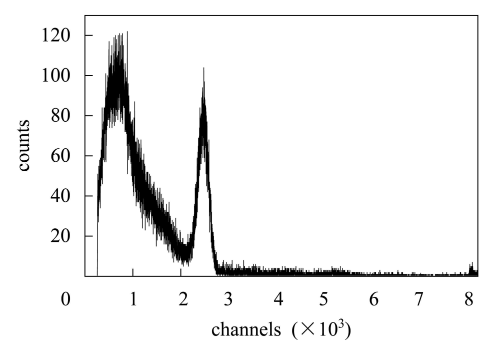

Fig. 1. Gamma ray spectra of ^{137}Cs (NaI).

In the convolution method, some math filters, such as [−3,12,17,12, −3]/35 for five-point smoothing, [−2,3,6,7,3,−2]/21 for seven-point smoothing described by Savitsky and Golay[1], Gaussian smoothing function and Lorentzian smoothing function described by Evans and Hiorns[10], are widely used to eliminate fluctuation[19]. Better result will be obtained while the width of math filter is approximated

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to the average FWHM (in channels) of the peaks in the spectra to be smoothed. However, this better result is based on the fact that fluctuation $N(x)$ and spectra $S(x)$ have different frequency and in the practical application, it is quite difficult to decide the average FWHM before calculating. Sophisticated value used in the processing always gives uncompleted or distorted elimination and cannot be fit for different spectra.

Recently, smoothing by spline function is used widely, because it leads to a very simple algorithm for the construction of the function and gives in general satisfactory results. In this paper, the least square method with Cubic B-spline basis functions is introduced to fit the gamma ray spectra without reprocessing. The description of this method and its application is divided into four sections. In Section 2, the B-spline basis functions and least square are described briefly. In Section 3, the noisy data are used to test this method with some discussion. Conclusions will be given in the last section.

2 Description of method

The Cubic B-spline basis functions is defined as a piecewise function which is non-zero only over four adjacent intervals between knots,

$$\phi(x)=\begin{cases}0, & |x|\geqslant 2\\ \frac{1}{2}|x|^3-|x|^2+\frac{2}{3}, & |x|\leqslant 1\\ -\frac{1}{6}|x|^3+|x|^2-2|x|+\frac{4}{3}, & 1<|x|<2\end{cases} \tag{1}$$

Because it is a spline and therefore has the appropriate continuity at given knots, its value of the first and second derivatives are zero at two end knots[20].

To define the full set of cubic B-splines which are required for our purpose in the range of interest channels $a\leqslant x\leqslant b$, where x is the channel number, firstly, it is necessary to select five channels x_0,x_1,x_2,x_3,x_4 with equal interval that satisfy $a\leqslant x_0<x_1<x_2<x_3<x_4\leqslant b$. With this set of channels as center knots, we define, as above, the fundamental splines $\phi_j(x)$ respectively, as shown in Fig. 2, where $j=0,1,\cdots,4$. Then the general cubic B-splines fitting curve with channels x_0,x_1,x_2,x_3,x_4 has the unique representation in the range $a\leqslant x\leqslant b$ of the form

$$S(x)=\sum_{j=0}^{4}c_j\phi_j\ . \tag{2}$$

The method of least square assumes that the best-fit $S^*(x)$ curve with selected channels has the minimal sum of the least square error from the measured spectra $F(x)$.

$$\|\delta_i\|^2=\sum_{x=1}^{m}[S_i^*(x)-F(x)]^2=\min\sum_{x=1}^{m}[S_i(x)-F(x)]^2\,, \tag{3}$$

$$S_i(x)=C_{i,0}\phi_0(x)+C_{i,1}\phi_1(x)+\cdots+C_{i,n}\phi_n(x),\ n\leqslant m. \tag{4}$$

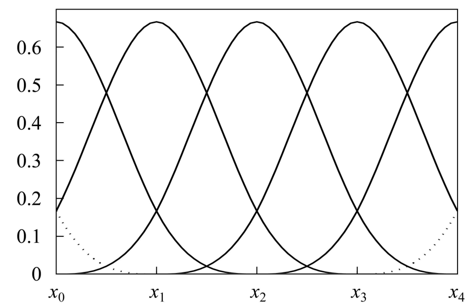

Fig. 2. The 5-fundamental splines (c_i=1).

Where m is the number of the channels of the spectra in the interval $[a,b]$, and n is the number of the selected channels. Then, new channels are selected as knots in the middle of each interval and the process is repeated until the interval is equal to 1 in which the fitting curve is the same as the measured spectra $F(x)$. Using the least square method, the best-fit function $S_i^*(x)$ with selected channels will be obtained at each time, which can form a set of fitting curves about the measured spectra $F(x)$. With this set of curves, however, as described by Naoki Saitou[21], the criterion of choosing the curve that gets the minimal $\|\delta_i\|^2$ as resultant curve is not exact, since this value is to decrease to zero as the number of knots increases and the minimum value of $\|\delta_i\|^2$ gives the interpolated value, that is, no smoothing effect is obtained.

With the finite set, comparing two adjacent fitting curves $S_i^*(x)$ and $S_{i+1}^*(x)$, the Residual Sum of Square (RSS) is calculated

$$\|\epsilon_i\|^2=\sum_{x=1}^{m}\left[S_{i+1}^*(x)-S_i^*(x)\right]^2, \tag{5}$$

which can be considered as the bias error of $S_i^*(x)$ relative to $S_{i+1}^*(x)$. The lower this value, the closer these two adjacent curves. This value can also indicate that the curve $S_{i+1}^*(x)$ contains more complex components than $S_i^*(x)$ which can be considered to have more noise in $S_{i+1}^*(x)$. Since the noise in the Fourier domain is in frequency order, hence, $S_i^*(x)$ can represent the measured spectra approximately if $\|\epsilon_i\|^2$ gets minimal value. However, as the number of selected channels increases, we find that the number of minimal value in the set of $\|\epsilon_i\|^2$ is not unique, sometimes, that is, some fitting curves are all satisfied with this condition. In this case, according to

Reinsch[22], the smoothest spline curve is characterized in such a way as to obtain the minimum value of $\int_a^b [(S_i^*)''(x)]^2 \mathrm{d}x$ in the interval $[a,b]$. Calculating the value of $\int_a^b [(S_j^*)''(x)]^2 \mathrm{d}x$ where $\|\epsilon_j\|^2$ gets the minimum value in the set of $\|\epsilon_i\|^2$, the resultant curve can be chosen according to the minimum value of $\|\epsilon_j\|^2$ and $\int_a^b [(S_j^*)''(x)]^2 \mathrm{d}x$.

3 Test and discussion

As a test of this method, some noisy data (both synthetic and experimental) are investigated in this section. Synthetic noisy data S using two gaussian functions with $\sigma_1^2 = 12$ and $\sigma_2^2 = 10$ is shown as the solid line in Fig. 3(a). The data are sampled at 129 evenly-spaced points in the given interval with the superimposed random noise varied as a function of the square root of the counts per channel.

According to the method described above, the values of $\|\epsilon_i\|^2$ are calculated as shown in Fig. 3(b). Because the selected channel number is 5 in the first fitting compared with 129 in the last fitting, the range of $\|\epsilon_i\|^2$ is from $\|\epsilon_2\|^2$ to $\|\epsilon_6\|^2$ which corresponds to the channels selected $(2^{i+1}+1)$.

In this sub-figure, the results clearly indicate that the minimum value is unique where $i = 4$ in the set of $\|\epsilon_i\|^2$, which means that the curve $S_4^*(x)$ can approximate to the noisy data well. The fitting result $S_4^*(x)$ is also shown in Fig. 3(a), respectively. The residual between the fitting curve using the cubic B spline basis function and the noise-free curve is shown in Fig. 3(c). It can be seen that the residual is mild and less than ±1 in most cases, that is, the fitting curve has a small difference with the noise-free curve that can estimate the noise-free curve approximately.

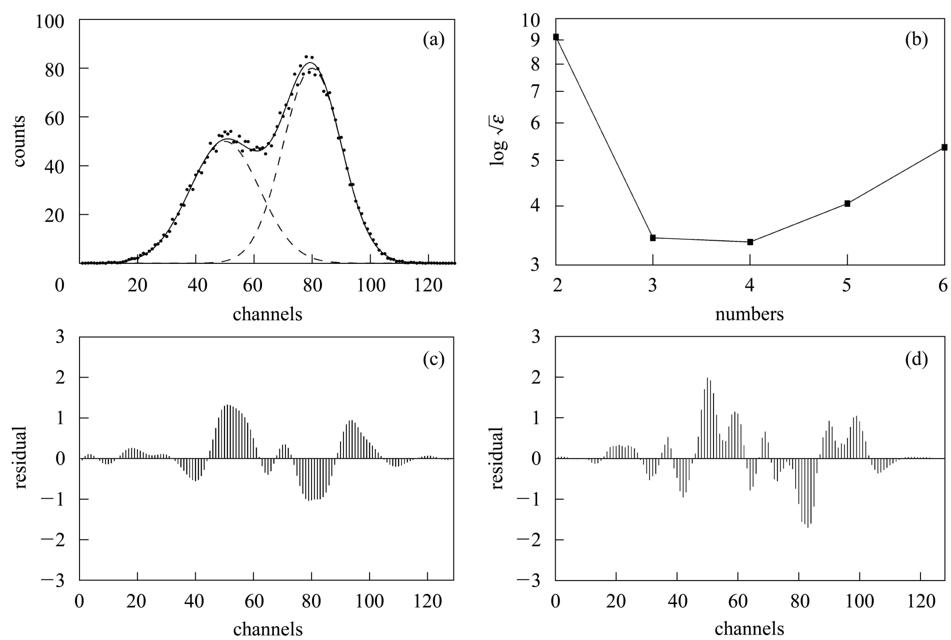

Fig. 3. Synthetic noisy data fitting.
(a) Synthetic noisy data, original curve, B-spline function fitting curve and convolution curve; (b) The logarithm of $\|\epsilon_i\|^2$ with i from 2 to 6; (c) The residual of cubic B-spline fitting curve with noise-free curve; (d) The residual of convolution method curve with noise-free curve.

Compared with this method, the noisy data are also processed by the convolution method with 9-point width filter estimated according to the value of σ. The result is shown in Fig. 3(a) with the residual shown in Fig. 3(d). As can be seen from the residual obtained by the convolution method, the difference is shaped shrilly and more than ±1 in most cases compared with mild shape and less than ±1 in most case while using the cubic B-spline method. It means that, the fitting curve obtained by the cubic B-spline method can give a more approximation to the noise-free curve than the curve obtained by the convolution method.

We then examine the effect of smoothing. In general, the elegant way to characterize the effect of smoothing of a curve is to analyze its spectrum in the Fourier domain. The curve with better smoothing effect will reserve the values for the Fourier coefficients corresponding to the low frequencies and get lower values for the high frequencies coefficients. Hence, in order to compare the effect of smoothing, the Fourier Transform result curves by the cubic B-spline method and the convolution method are obtained as shown in Fig. 4. It can be seen from this figure, the low frequency components are reserved satisfactorily with both the cubic B-spline method and the convolution method(below coefficient 8). However, the convolution method becomes helpless in the case that the noise and noise-free curves have approximate frequency, as shown in the range from 8 to 10. In this range, the noise components can not be reduced with the convolution method which may appear as ripples in the curve. By contrast with this, the cubic B-spline method can remove the noise components sufficiently. In the middle frequency region between 10 and 25, the Fourier amplitude value of the cubic B-spline method approaches more to the amplitude value of noise-free than that of the convolution method, that is, the noise components in the range of these frequencies, are removed more sufficiently by the cubic B-spline method. With the increase of frequency, this value varies slightly and keeps a lesser value with the cubic B-spline method, which gives a smoother profile of the fitting curve.

The most important characteristic of the method described in this paper is that it can obtain the best fitting curve and determine the smoothing end condition automatically without any initial input value. The least square method described in this paper, gives the fitting curve minimal sum of the least square error from the noisy data at each time and with the increase of channels selected, all the fitting curves obtained by the least square with the cubic B-spline basis function form a finite set. Anyone from this set is the best fitting curve corresponding to the selected channels and the result curve determined with the criterion described as above is chosen from this set, so the result curve is the best fitting curve.

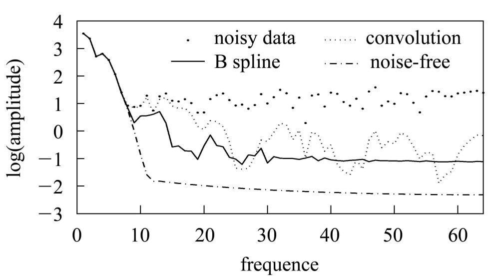

Fig. 4. The Fourier transform result curves obtained by the cubic B-spline method and the convolution method.

Herein, as described by Naoki Saitou[21], we don't recommend the minimum RSS of fitting curve with noisy data as the criterion to determine the best fitting curve, because RSS decreases to zero as the number of selected channels increase and the minimum value gives interpolated result without smoothing effect.

The criterion used in this paper is based on the idea of Reinsch[22]. In order to show the method clearly, the Reinsch's criterion is described briefly as follows. In the interval $[a,b]$ with m channels, the smoothing function $S(x)$ to be constructed shall minimize the value of $\int_a^b [(S^*(x))''(x)]^2 \mathrm{d}x$ among all functions $S^*(x)$ such that

$$\sum_{x=1}^{m}\left[\frac{S^*(x)-F(x)}{W_x}\right]^2 \leqslant C \ ,$$

where W_x is the weight assigned to channel x, and C is a smoothing parameter which controls the extent of smoothing. When $C = 0$, no smoothing is carried out and the noisy data are interpolated and as C increases, the degree of smoothing increases. However, it should be noted that this condition has little meaning in the noise elimination especially in the reduction of statistical noise, since the degree of smoothing required in the statistical noise elimination of spectra is far greater than that is normally required and the values of C and W_x are difficult to be determined before calculating.

To avoid these problems, RSS of two adjacent fitting curves $S_i^*(x)$ and $S_{i+1}^*(x)$ are calculated. Since the noise in the Fourier domain is in frequency order and the fitting curves in the fitting set are also in order corresponding to the number of selected channels, the complex components contained in curve $S_{i+1}^*(x)$ can be considered to have more noise than $S_i^*(x)$. Minimum RSS means that these two adjacent curves have little difference and little noise $S_i^*(x)$ contained than $S_{i+1}^*(x)$ which can estimate noisy data approximately. However, with the fitting curve set, minimum RSS is not unique sometimes, that corresponds to more than one fitting curves. In this case, according to Reinsch's first condition that the smoothest spline curve has minimum value of $\int_a^b [(S_i^*)''(x)]^2 \mathrm{d}x$ in the interval $[a,b]$, the result curve is obtained. There is no initial input parameter to be determined before calculation while using the criterion described above and trivial calculations are also avoided.

Another praiseworthy characteristic is that, the cubic B-spline basis function fitting method can give an explicit expression of the result curve that is convenient for calculating its derivative and area.

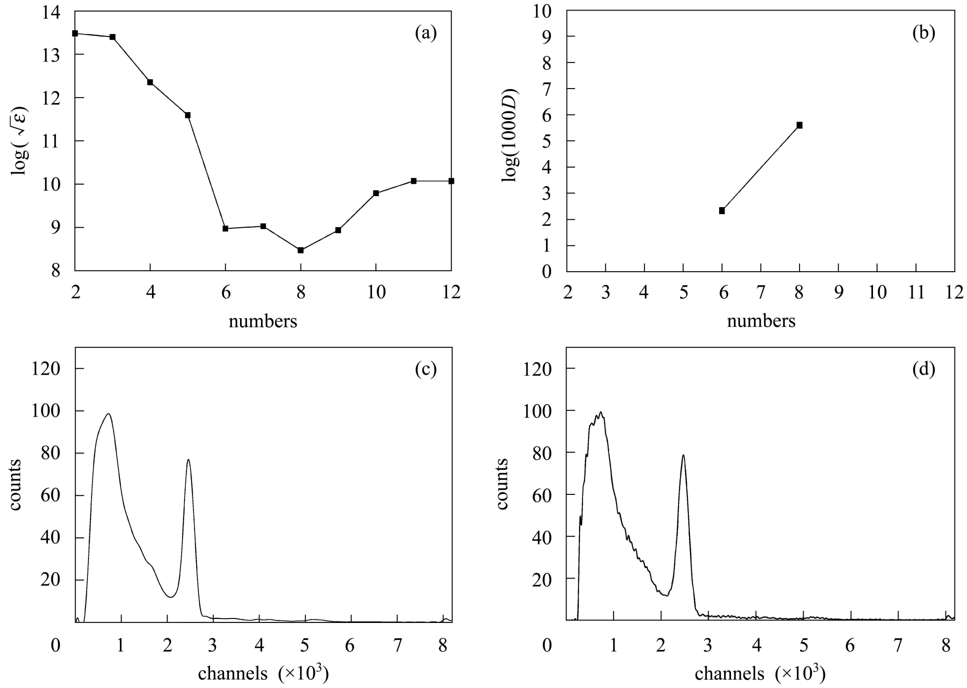

Fig. 5. γ-ray spectrum of ^{137}Cs (NaI) and its fitting result using B-spline basis functions and the convolution method. (a) The logarithm of $\|\epsilon_i\|^2$ with i from 2 to 12; (b) The square integral values of $(S_6^*)''(x)$ and $(S_8^*)''(x)$; (c) The fitting curve $S_6^*(x)$ with the cubic B-spline basis function; (d) The fitting curve $S_8^*(x)$ with the cubic B-spline basis function.

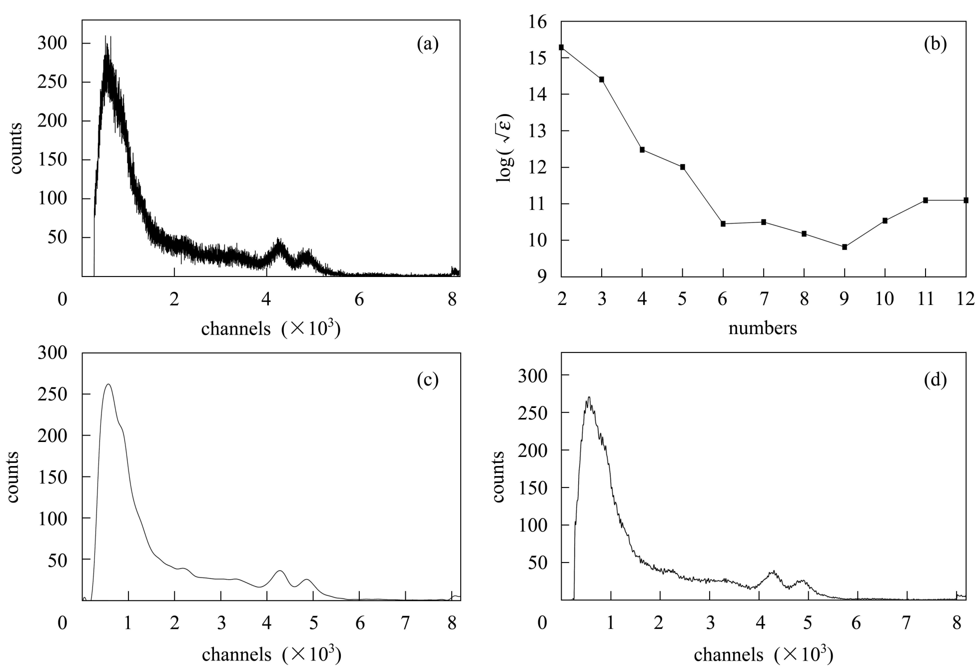

Fig. 6. Gamma-ray spectrum of ^{60}Co (NaI) and its fitting result using B-spline basis functions. (a) The experimental spectra of ^{60}Co source; (b) The logarithm of $\|\epsilon_i\|^2$ with i from 2 to 12; (c) The fitting curve with $i = 6$; (d) The fitting curve with $i = 9$.

Table 1. Least square error of corresponding best-fit curves.

source	channels	N^*	$\|\epsilon_2\|^2$	$\|\epsilon_3\|^2$	$\|\epsilon_4\|^2$	$\|\epsilon_5\|^2$	$\|\epsilon_6\|^2$	$\|\epsilon_7\|^2$	$\|\epsilon_8\|^2$	$\|\epsilon_9\|^2$	$\|\epsilon_{10}\|^2$	$\|\epsilon_{11}\|^2$	$\|\epsilon_{12}\|^2$
^{137}Cs	8193	6	0.7167	0.6595	0.2324	0.1084	0.0079	0.0083	0.0048	0.0076	0.0178	0.0237	0.0238
^{60}Co	8193	7	0.4346	0.1802	0.0263	0.0164	0.0035	0.0036	0.0026	0.0018	0.0038	0.0066	0.0066
mixture	8193	5	0.0038	0.0061	0.0057	0.0009	0.0001	0.0002	0.0004	0.0008	0.0017	7.1099	7.1099

N^* is magnitude of $\|\epsilon_i\|^2$.

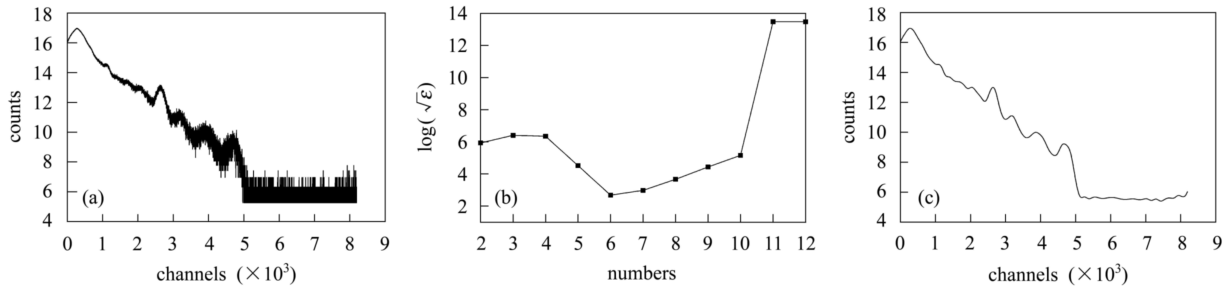

Fig. 7. Mixed radiate source and its fitting result. (a) The experimental spectra of mixture source; (b) The logarithm of $\|\epsilon_i\|^2$ with i from 2 to 12; (c) The fitting curve with $i = 6$.

The experimental data shown in Fig. 1 are measured from ^{137}Cs source with 8193 channels by NaI detector for more than 2 hours. According to the method described in this paper, the values of $\|\epsilon_i\|^2$ are calculated as shown in Fig. 5(a) with detailed value listed in Table 1. Because the selected channel number is 5 in the first fitting compared with 8193 in the last fitting, the range of $\|\epsilon_i\|^2$ is from $\|\epsilon_2\|^2$ to $\|\epsilon_{12}\|^2$. What can be seen from this sub-figure is that two minimum values about $\|\epsilon\|^2$ satisfy the condition where i=6 and i=8 in the fitting set. The curves $S_6^*(x)$ corresponding to the minimum values is shown in Fig. 5(c) as well as the values of $\int_1^{8193} [(S_6^*)''(x)]^2 \mathrm{d}x$ and $\int_1^{8193} [(S_8^*)''(x)]^2 \mathrm{d}x$ are shown in Fig. 5(b) which can indicate the smoother fitting curve clearly.

By comparison with this method, the spectrum is also smoothed by the 3-point convolution method(iterates 2000). However some ripples and somewhat distortion are appeared in the smoothed curve. As a result, the cubic B-spline method can eliminate fluctuation noise sufficiently with little distortion and the result fitting curve is smoother. This is of great significance in peak searching, especially in distinguishing the overlapping peaks.

The gamma ray spectrum of ^{60}Co and the mixed radiate source are also measured in the same environment to test this method, as shown in Fig. 6(a) and Fig. 7(a), respectively. The values of $\|\epsilon_i\|^2$ are listed in Table 1, also shown in Fig. 6(b) and Fig. 7(b). The difference between them is that there are two minimum $\|\epsilon_i\|^2$ values about ^{60}Co compared with the unique value of the mixture source. The fitting curves corresponding to the minimum value of $\|\epsilon_i\|^2$ are shown respectively in each figure. Because of two minimum of $\|\epsilon_i\|^2$ in ^{60}Co source, the values of $\int_1^{8193} [(S_i^*)''(x)]^2 \mathrm{d}x$ should be compared to determine the smoother curve as the result. In the calculation of mixed source, the fitting curve corresponding to the unique value of $\|\epsilon_i\|^2$ is best.

4 Conclusion

In the paper, a least square fitting method with the cubic B-splines basis functions has been developed for eliminating the statistical fluctuation of gamma-ray spectra. As one can see from the illustration as above, the method is automatic and does not need any initial input value. Different tests show that this method can remove statistical fluctuation sufficiently with little data distortion and the resultant fitting curve is smoother than that obtained by the convolution method.

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