Statistical distribution based detector response function of a Si(PIN) detector for K_α and K_β X-ray*

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Abstract: A semi-empirical detector response function (DRF) model of a Si (PIN) detector is proposed to fit element K_α and K_β X-ray spectra, which is based on the statistical distribution analytic (SDA) method. The model for each single peak contains a step function, a Gaussian function, and an exponential tail function. Parameters in the model are obtained by the weighted nonlinear least-squares fitting method. In the application, six kinds of elements' characteristic X-ray spectra are obtained by using the Si (PIN) detector, and fitted by the established DRF model. Reduced chi-square values are at the interval of 1.11–1.25. Other applications of the method are also discussed.

Key words: detector response function, Si (PIN) detector, statistical distribution analytic method, weighted nonlinear least-squares fitting

PACS: 29.30.Kv, 29.85.fj **DOI:** 10.1088/1674-1137/37/1/018202

1 Introduction

An accurate analytical description of photopeaks continues to be an important facet of X-ray spectroscopy. Precise line shape representation is one of the essential parts of accurate energy and intensity determination in X-ray spectrum analysis. Much research work has dealt with analytic functions for gamma and X-ray spectrum analysis by the least-square method [1–5]. One purpose of them is to outline a method to generate the response function of the detector. The main portion of the line shape should not be Gaussian but rather Voigtian since the observed shape is the convolution of the Gaussian with the intrinsic Lorentzian profile. However, for K X-rays of low and medium Z elements and L X-rays of medium and high Z elements, the intrinsic X-ray linewidth is only a few eV. Since it is small compared with the typical full width at half maximum of a silicon detector (100–300 eV), the Lorentzian convolution is generally omitted and Gaussian based functions are accepted as an adequate representation of an X-ray line shape [6]. In the present work, the photopeak can be expressed as a simple Gaussian distribution. The Gaussian distribution of discrete energy peaks, which was established by researchers from the Center for Engineering Applications of Radioisotopes (CEAR) has been accepted by most researchers in this area [7, 8].

Previous studies have shown that X-ray spectra from silicon detectors contain four or five compositions [9–12]. Parameters of the model were obtained from curve fitting and were expressed as functions of the X-ray energies. All of them were based on two parts: the theory of the interaction between photon and materials; the mathematical proximity guess to fit the energy peak. The previous research works mentioned above all contain a Gaussian function with the basic format:

$$G(i)=A\exp\left[\frac{-(i-i_0)^2}{2\sigma^2}\right]. \quad (1)$$

Parameters can be obtained by fitting serial experimental full energy peaks, and this function can reflect the statistical nature of system noise and the charge collection process.

Having studied and compared other detector response function (DRF) compositions for Si(Li), we proposed an improved DRF for our vacuum system with

Received 2 March 2012, Revised 16 August 2012
∗ Supported by National Natural Science Foundation of China (40974065, 41025015), Scientific and Technological Innovative Team in Sichuan Province (2011JTD0013)and "863" Program of China (2012AA063501)

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three parts which can describe the spectra from Si(PIN) detector more appropriately for detected elements K_α and K_β line characteristic X-ray spectra. A statistical distribution-based analytic (SDA) theory was presented to implement the DRF model.

2 Analytical fitting model of K X-ray spectrum

2.1 The choice of detector response function forms

For our Si(PIN) detector based energy dispersive X-ray fluorescence (EDXRF) system, we established a new DRF preliminarily to analyze some major elements of vanadium titanomagnetite, such as Ti, V, Fe, Ni, Cu, and Zn. The Si escape peak was ignored in our detecting system [13–15], as it was very weak and out of the analysis range. So the response function model $R(E_i)$, similar to that Campbell J L et al. proposed [16], has the following three functions: (1) a step function on the low energy side of full energy peak, $S(E_i)$; (2) a Gaussian function of full energy peak, $G(E_i)$ and (3) an exponential tail function on low energy side of full energy peak, $T(E_i)$. The expressions are

$$S(E_i) = H_S \cdot \pi \cdot \mathrm{erfc}\left(\frac{E_i - E_k}{\sqrt{2}\cdot\sigma_k}\right), \tag{2}$$

$$G(E_i) = H_G \exp\left(-\frac{(E_i - E_k)^2}{2\sigma_k^2}\right), \tag{3}$$

$$T(E_i) = H_T \cdot \exp\left(\frac{E_i - E_k}{\beta}\right) \cdot \mathrm{erfc}\left[\frac{E_i - E_k}{\sqrt{2}\sigma_k} + \frac{\sigma_k}{\sqrt{2}\beta}\right], \tag{4}$$

$$R(E_i) = S(E_i) + G(E_i) + T(E_i), \tag{5}$$

where i is the channel number, E_k is the incident photon energy, E_i is the portion of that energy deposited in the detector, H_S, H_G, H_T are free parameters of the background, the Gaussian and exponential tail, σ_k is the standard deviation of full energy peak and β is the slope of the exponential feature. Parameters of H_S, H_G, H_T, β and σ_k are obtained by using the weighted nonlinear least squares fitting method [17]:

$$\min \sum_i w_i (N(E_i) - R(E_i))^2, \tag{6}$$

where w_i is the weight for E_i, approximate to the reciprocal of the variance of $N(E_i)$; $N(E_i)$ is the counts in channel i; $R(E_i)$ is the fitting value at channel i.

The interaction of X-rays with the detector crystal obeys a certain statistical fluctuation. So the detector response function can be formulated as a statistical distribution-based analytic (SDA) model, which is similar to the probability distribution function (pdf) [18].

If $R(E_i)$ is to be a statistical distribution-based analytic (SDA) model, it must meet two requirements:

$$\begin{cases} R(E_i) \geqslant 0 (\mathrm{keV}^{-1}) \\ \int R(E_i) \mathrm{d}E_i = 1 \end{cases}, \quad -\infty < E_i < \infty. \tag{7}$$

In practice, E_i is greater than or equal to zero, usually within a given range. Linear parameters (denoted as H_S, H_G, and H_T) are the normalized response function $R(E_i)$.

2.2 Gaussian-shaped peak standard deviation

The use of the Gaussian distribution of discrete energy peaks is accepted by most researchers in this field. In the former reported studies, the standard deviation of the Gaussian part of full energy peak can be fitted to

$$\sigma_k = \sqrt{\sigma_e^2 + \varepsilon F E} \ \mathrm{keV}, \tag{8}$$

where ε is the energy to create one electron-hole pair for Si, F is the Fano factor, E is the incident photon energy and σ_e is the electronic noise.

In this paper, the values of σ_k for different energies are fitted by using the weighted least squares fitting method based on SDA theory, and then σ_e can be also fitted by using least-squares regression based on Eq. (8). The fitting function of σ_k and E (4–25 keV) for this Si-PIN detector is

$$\sigma_k = \sqrt{0.00386 + 0.00111 \cdot E} \ \ \mathrm{keV}. \tag{9}$$

As compared to the function in Ref. [19], which was

$$\sigma_k = \sqrt{0.00136 + 0.0006335 \cdot E} \ \ \mathrm{keV}. \tag{10}$$

The detector used in Ref. [19] was a liquid nitrogen-cooled Si(Li) semiconductor X-ray radiation detector, and it has better energy resolution than a Si-PIN detector, about 158 eV FWHM at the 5.9 keV Mn K_α line. A Si-PIN detector is different to a Si(Li) detector (Li-drifted) in its structure and composition. As compared to a Si(Li) detector, the electronic noise of a Si-PIN detector is still much larger, because its capacitance increasing with the increase of effective detector area, but its detection efficiency is also increasing which can bring a good statistic effect. X-rays interact with silicon atoms to create an average of one electron/hole pair for every 3.73±0.09 eV of energy lost in the Si p-i-n diode between 80 and 270 K [20]; the Fano factor for a Si-PIN semiconductor detector ranges from 0.1 to 0.13. Resolution depends not only on the detector but also the electronics.

In practice, σ_k for each full-energy peak can be calculated by the SDA method after measuring several pure-element X-ray pulse-height spectra. If the experimen-

tal condition is stable enough, such a measurement only needs to be implemented once for all related elements.

3 Experiment and results analysis

3.1 Experimental procedure

An EDXRF system, series CIT-3000SM, developed by ourselves was employed to implement the experiment [21]. The energy resolution of the Si-PIN detector in this X-ray analyzer is 220 eV@ 5.9 keV (^{55}Fe). The whole EDXRF system was integrated in a vacuum house containing a vacuum chamber, an X-ray tube, a sample carrier, and a Si (PIN) detector; samples were measured under vacuum conditions of about −0.095 MPa because the count rate of X-ray spectra detected under vacuum conditions was higher and the background was lower than those detected under atmospheric pressure. Meanwhile, a filter was also fixed to reduce the background and Compton scattering. Six kinds of chemical analytical reagents, TiO_2, V_2O_5, Fe_2O_3, Ni powder, CuO, and ZnO, were prepared and measured separately by the EDXRF system, which employed an X-ray tube to excite the element's characteristic X-rays and a Si (PIN) detector to detect characteristic X-rays.

In this study, the appropriateness of a particular function was determined by the visual agreement between fitted function and experimental data for the fitting, and by the reduced chi-square value (χ_r^2).

$$\chi_r^2=\frac{1}{M}\sum_{i=l}^{r}\frac{(N(E_i)-R(E_i))^2}{N(E_i)}, \tag{11}$$

where l and r are the channels specifying the fitting interval, i is the channel number. M is the number of degrees of freedom, $M=r-l+1-2\times f$, f is the number of variable parameters in the functional form fitted to one peak, there are four in total: H_S, H_G, H_T, and β; 2 represents the number of space line for each element, K_α and K_β are taken into account in this paper.

Measured data and fitting results were recorded in Table 1, fitting to an element K X-ray doublet in the Si(PIN) detector is shown in Fig. 1. A relatively good fit is indicated by the minimal value of χ_r^2. The spectrum fraction (SF) for each peak was also achieved.

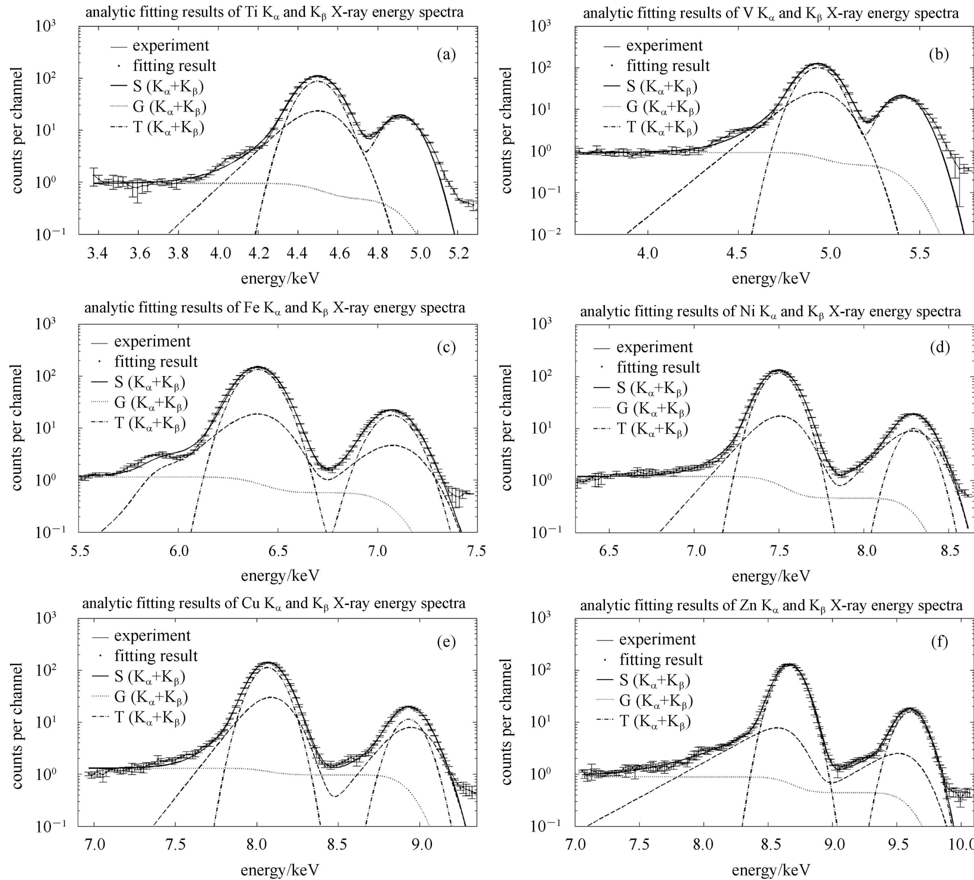

Fig. 1. Fits to Ti, V, Fe, Ni, Cu, Zn K X-ray doublets in Si (PIN) detector. In these six subfigures each component for K_α and K_β is S+G+T except the K_β spectra in (a) and (b) which are S+G. An escape peak of Si was considered and fitted in Fe spectrum at the low energy part of K_α.

Table 1. The measured data and fitting results of six element K X-ray doublet full energy peak spectrum[a)].

	Ti	V	Fe	Ni	Cu	Zn
E_{K_α}	4.4989	4.9381	6.4006	7.498	8.0644	8.6662
σ_{K_α}	0.0941	0.0967	0.1047	0.1104	0.1132	0.1161
(k_α,n)	(188,11)	(206,11)	(275,15)	(337,18)	(369,17)	(403,19)
E_{K_β}	4.9137	5.4017	7.0732	8.2945	8.9317	9.6043
σ_{K_β}	0.0965	0.0993	0.1082	0.1143	0.1174	0.1205
(k_β,n)	(205,9)	(225,9)	(313,15)	(382,14)	(418,14)	(456,14)
χ_r^2	1.2263	1.2512	1.1786	1.1064	1.1150	1.1063
$E_l{-}E_r$ (M)	3.3765–5.2797 (73)	3.5717–5.7677 (85)	5.4802–7.4626 (105)	6.3121–8.6131 (123)	6.9670–9.3388 (127)	7.0555–10.645 (163)
[b)]SF	6.6653	6.5218	7.4771	7.469	7.9373	8.4266

a) The units of E_{K_α}, σ_{K_α}, E_{K_β}, σ_{K_β}, E_l, E_r in Table 1 are keV. b) SF is the spectrum fraction of K_α characteristic X-ray full energy peak area to K_β's.

In Table 1, k_α and k_β indicate the channel of full energy peak center for K_α and K_β X-ray respectively, and values of n indicate the range of measured full energy peak from $k{-}n$ to $k{+}n$. The values of σ_{K_α} and σ_{K_β} were calculated under this range, respectively. The χ_r^2 value of each element's K X-ray doublets spectra was calculated out with the degree of freedom M corresponding to the range $E_l{-}E_r$.

The DRF model based on the SDA method fitted six medium atom elements' K_α and K_β X-ray pulse-height spectrum of Si(PIN) detector with χ_r^2 values closed to 1, from 1.11 to 1.25. The main part of each peak was a Gaussian function, and the Gaussian-shaped part could be used to calculate net peak area. The other two parts of DRF model reflected the background and electronic noise, etc. K_β spectra of Ti and V were overlapped with their K_α spectra, judged by the interaction between $(E_{K_\alpha}{-}3\sigma_{K_\alpha}, E_{K_\alpha}+3\sigma_{K_\alpha})$ and $(E_{K_\beta}{-}3\sigma_{K_\beta}, E_{K_\beta}+3\sigma_{K_\beta})$, which can be considered as a standard judgment. The background and Compton scattering were controlled well, as shown in energy spectra, but the S part in DRF could not be ignored because the background and Compton scattering still existed. As shown in Fig. 1, much better fitting results can be achieved at the low energy part of each spectrum than that at high energy part especially beyond the K_β peak. Other studies [22–24] found a similar feature to this paper. Because the detector response function mainly describes the full energy peak and its contribution to the low energy part of the peak, so DRF models could not fit the high energy range well enough.

3.2 Standard deviation of Si(PIN)

The Si(Li) detector and Si(PIN) detector are both silicon detectors, and many previous studies have focused on the Si(Li) detector. In this study, the standard deviation of a Si(PIN) detector is also taken into account to analyze the detector resolution and electronic feature. The electronic noise σ_e in Eq. (8) was a free parameter of the model. Parameters in Eq. (8) were fitted and compared with others. Parameters for Si(Li) obtained by other studies and for Si(PIN) obtained by this paper are shown in Table 2.

As shown in Table 2, the deduced electronic noise FWHM is 62.13 eV, and the Fano factor F is 0.292 for Si-PIN detector. The value of the Fano factor is larger than most values in the literature, probably because the Si-PIN detector we used is worked under 233.15 K (not 77 K in normal), and our fitting values extend to 25 keV where the Si-PIN detector resolution is poor. The mean impact ionization energy ε was assumed to be 3.8 eV for the Si(PIN) operating at 233.15 K [26]. The main differences of Si(Li) and Si(PIN) detectors are the values of electronic noise. As a Si(PIN) detector works using electronic cooling; its energy resolution (FWHM) is not as good as a Si(Li) detector, such as for ^{55}Fe, the FWHM is 150–180 eV for Si(Li), about 200–240 keV for Si(PIN). Generally, an experimental EDXRF instrument is stable enough; the value of σ can be considered as a constant parameter for each element's characteristic X-ray.

Table 2. Parameters in Eq. (8) of Si detectors from different research works.

method	parameter value
T. He, et al. [19]	Si(Li): σ_e=36.88 eV, ε=3.793 eV, F=0.167
Yosuke INAGAKI, et al. [25]	Si(Li): σ_e=44.5 eV, ε=3.76 eV, F=0.127
F. Scholze, et al. [10]	Si(Li): [a)]ΔE_{el}=44 eV, ε=3.7eV, F=0.114
this paper	Si(PIN): [b)]σ_e=62.13 eV, ε=3.796 eV, F=0.292

a) In Scholze's study, ΔE_{el} represents to the electronic noise. b) σ_e and F are obtained by Eq. (8) and Eq. (9)

4 Conclusion

The DRF model for the Gaussian-shaped full energy peak of the detected X-ray pulse-height spectrum can be formulated as a probability distribution function. The SDA fitting method is not the same as other traditional peak analytic methods, as it can fit the two K line X-ray peak simultaneously. Parameter values of K line doublet X-ray peaks are obtained by using the weighted nonlinear least squares fitting method, and parameters of the full energy peak will not change if the experimental condition is invariant. This characteristic made SDA a relatively simple and commonly used technique to carry out radiation spectrum analysis procedures. χ_r^2 values of K line doublet X-ray peaks detected by the Si (PIN) detector for each element were in the range of 1.11 to 1.25. The established DRF model can be also used to obtain the detector response function for many applications involving X-ray spectroscopy, or to broaden a simulated X-ray pulse-height spectrum when using the Monte Carlo simulation approach. This is being investigated and will be reported later.

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