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Improved peak shape fitting in alpha spectra

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HIGHLIGHTS

- A new algorithm for analysis of alpha-particle spectra is presented.
- The peak shape functions reproduce low- and high-energy tailing.
- Its proficiency is demonstrated by a fit to high-resolution spectra.
- It outperforms existing specialised fit functions.

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ABSTRACT

Peak overlap is a recurrent issue in alpha-particle spectrometry, not only in routine analyses but also in the high-resolution spectra from which reference values for alpha emission probabilities are derived. In this work, improved peak shape formulae are presented for the deconvolution of alpha-particle spectra. They have been implemented as fit functions in a spreadsheet application and optimum fit parameters were searched with built-in optimisation routines. Deconvolution results are shown for a few challenging spectra with high statistical precision. The algorithm outperforms the best available routines for high-resolution spectrometry, which may facilitate a more reliable determination of alpha emission probabilities in the future. It is also applicable to alpha spectra with inferior energy resolution.

1. Introduction

Spectral deconvolution has always been an issue in alpha-particle spectrometry (García-Toraño, 2006; Pommé, in press), as individual alpha peaks tend to partially overlap due to the limited attainable energy resolution, even in ideal conditions. The benefit of spectral analysis is the identification and separation of peaks, with the aim of determining activity ratios between different alpha emitters or calculating individual emission probabilities in the decay of a single radionuclide. In particular for the latter application, i.e. deriving reference decay data from high-resolution spectra, spectral deconvolution is performed by fitting complex mathematical functions to each peak. The normalised area of each fitted peak in the energy spectrum then represents its relative emission probability. The quality of the emission data depends on the realistic representation of the peak shape and the ability to closely reproduce the experimental energy spectrum.

Various peak models have been proposed in the past (Trivedi, 1969; Baba, 1978; Wätzig and Westmeier, 1978; García-Toraño and Aceña, 1981; L'Hoir, 1984; Bortels and Collaers, 1987; Koskelo et al., 1996; García-Toraño, 1997, 2003; Lozano et al., 2000; Pommé and Sibbens, 2008; Semkow et al., 2010) and in recent work an attempt was made to include more physics in spectrum deconvolution (Siiskonen and Pöllänen, 2011; Westmeier and Siemon, 2012; Pöllänen and Siiskonen, 2014) or to use an artificial neural network (Baeza et al., 2011). One of the most successful analytical models to represent the shape of a mono-energetic alpha peak is the convolution of an exponential low-energy tail with a Gaussian distribution, as suggested by L'Hoir (1984) and introduced by Bortels and Collaers (1987)

$$f(u-\mu;\sigma,\tau)=\frac{A}{2\tau}\exp\left(\frac{u-\mu}{\tau}+\frac{\sigma^2}{2\tau^2}\right)\operatorname{erfc}\left(\frac{1}{\sqrt{2}}\left(\frac{u-\mu}{\sigma}+\frac{\sigma}{\tau}\right)\right) \quad (1)$$

where A is the peak area, $u-\mu$ is the distance to the peak position, σ is the standard deviation of the Gaussian and τ is the tailing parameter. They obtained a better fit after pre-treating the spectrum by subtracting a long tail distribution and then fitting a mix

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of two exponential functions with different characteristic lengths, τ_1 and τ_2, using normalised weighting factors η_1 and $1-\eta_1$. This was implemented in the software package ALFA (Babeliowsky and Bortels, 1993). Pommé and Sibbens (2008) abolished the pre-treatment of the spectrum and added the third exponential function into the fit function with a normalised weighting factor $\eta_3=1-\eta_1-\eta_2$

$$F(u) = \sum_{i=1}^{3} \eta_i f(u-\mu_i;\, \sigma_i,\, \tau_i) \tag{2}$$

This function, implemented as visual basic application ALPHA in Excel©, has been applied in spectral deconvolution of ^{235}U (García–Toraño et al., 2005), ^{240}Pu (Sibbens and Pommé, 2004; Sibbens et al., 2010), ^{236,238}U (Marouli et al., 2014; Pommé et al., 2014) and the ^{230}U and ^{225}Ac decay series (Marouli et al., 2012; 2013). Whereas ALPHA generally performs very well, there are still noticeable residuals when fitting spectra that have millions of counts in each energy bin, since the extremely low random variations in the spectra reveal the slightest mismatch between fit and experiment. Similar imperfections show up with other analysis software, such as ALPACA (García-Toraño,1997) and ALFITEX (Caro Marroyo et al., 2013). Whereas ALFITEX applies the same peak model with the optional use of two or three exponentials (Eqs. (1) and (2)), ALPACA relies on a different peak shape model in which the low energy region is reproduced by a hyperbolic function.

A more adequate fit of the most challenging spectra requires more elaborate modelling. This adds to the complexity and computing time for reaching the optimum fit parameters via search routines. In this work, a logical extension to the analytical model in Eqs. (1) and (2) is proposed. The analytical model is implemented as a call function in an Excel spreadsheet and the built-in optimisation routine SOLVER is used to find the best match between fitted and measured spectrum. This spectral analysis tool will be referred to as 'BEST' and its performance will be tested on recently published ^{240}Pu and ^{238}U spectra and compared with existing models.

2. The 'BEST' algorithm

The typical line shape of a peak with triple tailing as defined in Eqs. (1) and (2) is shown in Fig. 1, with indications where the peak width σ and the tailing parameters $\tau_1<\tau_2<\tau_3$ determinate the peak shape. With increasing distance from the peak top, the tailing is dominated by exponentials with larger τ values. The analytical peak shape is not completely smooth in transition regions where the dominance changes from one exponential tail to another.

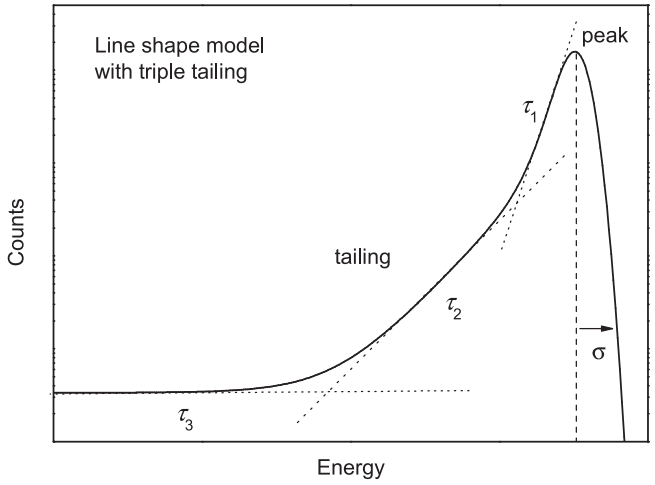

Fig. 1. Typical line shape applying a convolution of a Gaussian with three left-handed exponentials (Eqs. (1) and (2)) and indication where the peak width σ and the tailing parameters $\tau_1<\tau_2<\tau_3$ are the most influential.

One way of mitigating the abrupt changes in the slope of the tailing is adding more exponential tails with progressively increasing τ values. On the other hand, the experimental spectra are also not perfectly smooth and may show mild curvatures that are not well reproduced by monotonic functions. Therefore the fit would not only benefit from a higher number of exponential tailing functions, but also from flexibility in the weighting factors, allowing some of them to be negative or larger than 1 on condition that the sum of all weighting factors remains one.

Another observation is that alpha peaks show slight distortions and even some tailing at the high-energy side. This aspect can be taken into account by applying also right-handed exponential tailing in addition to the left-handed tailing. The inversion of the tailing is easily established by changing the sign of the position in the function call, i.e. by replacing $u-\mu$ with $\mu-u$. In the current implementation of the BEST algorithm, the peak shape can contain up to ten left-handed and four right-handed tailing functions

$$F(u) = \sum_{i=-3}^{0} \eta_i f(\mu_i-u;\, \sigma_i,\, \tau_i) + \sum_{i=1}^{10} \eta_i f(u-\mu_i;\, \sigma_i,\, \tau_i) \tag{3}$$

The sum of the weighting factors is normalised to one, but this is the only constraint on the individual values. In practice, this constraint is implemented by excluding η_1 from the list of free parameters in the fit and setting it equal to 1 minus the sum of the other factors

$$\sum_{i=-3}^{10} \eta_i = 1 \text{ or } \eta_1 = 1 - \sum_{\substack{i=-3 \\ i\neq 1}}^{10} \eta_i \tag{4}$$

In the decay of an alpha-emitting radionuclide, usually there are several alpha transitions involved with different energy and relative emission probabilities. Each of them corresponds to an individual peak in the alpha spectrum and the peak area A_k of peak k can be represented as the product of the total number of alpha decays A_{tot} of this particular nuclide multiplied by the relative intensity I_k

$$A_k = A_{\text{tot}} I_k = I_k \sum_{k=1}^{n_{peaks}} F_k(u) \tag{5}$$

It is convenient to use A_{tot} as a free fitting parameter to adjust all peaks of a radionuclide to the number of counts in a spectrum, without changing the spectral shape through the relative peak intensities. This feature can be used i.a. or e.g.. for determining the activity ratio of radionuclides in a mixed source. Some of the intensities may be released for more precise fitting, while others may be kept fixed, e.g. because literature values or information derived from gamma spectrometry may be more accurate. The normalisation of the intensities is enforced by constraining the intensity of the major peak to the value of 1 minus the sum of the intensities of the other peaks

$$\sum_{k=1}^{N_{peaks}} I_k = 1 \text{ or } I_1 = 1 - \sum_{k=2}^{N_{peaks}} I_k \tag{6}$$

Additional functional relationships between parameters, e.g. a constant ratio between emission probabilities, can be introduced as a constraint in the spreadsheet. Also the energy calibration, e.g. through a linear relationship between bin number and keV, can be part of the optimisation process, before individual peak positions are released in the fit for more precise positioning.

When fitting a spectrum within a selected energy region, all