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Spectrum unfolding in X-ray spectrometry using the maximum entropy method

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HIGHLIGHTS

- A new strategy to solve the unfolding problem in X-ray spectrometry is presented.
- The presented strategy uses a suitable combination of the codes MAXED and GRAVEL.
- The applied strategy provides additional information on the Detector Response Function.
- The code UMESTRAT is developed to apply this new strategy in a semi-automatic mode.

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ABSTRACT

The solution of the unfolding problem is an ever-present issue in X-ray spectrometry. The maximum entropy technique solves this problem by taking advantage of some known a priori physical information and by ensuring an outcome with only positive values. This method is implemented in MAXED (MAXimum Entropy Deconvolution), a software code contained in the package UMG (Unfolding with MAXED and GRAVEL) developed at PTB and distributed by NEA Data Bank. This package contains also the code GRAVEL (used to estimate the precision of the solution). This article introduces the new code UMESTRAT (Unfolding Maximum Entropy STRATegy) which applies a semi-automatic strategy to solve the unfolding problem by using a suitable combination of MAXED and GRAVEL for applications in X-ray spectrometry. Some examples of the use of UMESTRAT are shown, demonstrating its capability to remove detector artifacts from the measured spectrum consistently with the model used for the detector response function (DRF).

1. Introduction

It is well known that any experimental spectrum is modified by the detection system, causing a loss of information. The whole influence of the detector is quite complex to characterize. It is usually expressed as the convolution of different effects such as the radiation transport inside the detector, the energy resolution and all the contributions related to the signal formation and processing (Knoll, 2000). All these effects form the detector response function (DRF). To recover the original spectrum it is necessary to remove the detector influence, by solving the so-called inverse problem. Mathematically, any measurement I is related to the DRF $R(E)$ and to the original X-ray spectrum $f(E)$ through a convolution integral. The continuous equation can be discretized leading to the following linear equation system:

$$I_k+\varepsilon_k=\sum_{i=1}^{n} R_{i,k}f_i \quad \text{with} \quad k=1,\ldots,m \tag{1.1}$$

ε represents the (unknown) measurement error and m the number of discrete energy bins. This matrix equation is ill-conditioned and it is necessary to refer to a regularization technique to solve it. There exist different algorithms which can be used to solve the inverse problem (Hussein, 2011). Most of these are purely mathematical criteria and may lead to unphysical solutions, such as negative spectra. The maximum entropy unfolding technique solves the inverse problem by imposing a set of physical constraints, taking advantage of the known *a priori* information available and preserving positive values for the X-ray spectrum. These physical constraints are introduced through the so-called *Cross Entropy* equation

$$S=-\sum_{i=1}^{n}\left\{f_i\ln\left(\frac{f_i}{f_i^{DEF}}\right)+\left(f_i^{DEF}-f_i\right)\right\} \tag{1.2}$$

where the f_i^{DEF} distribution is the *default spectrum* which contains all the *a priori* information on the original spectrum. The *Cross Entropy* is

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always $S \leq 0$ for every f_i, and the condition $S=0$ is verified only when the original spectrum reconstruction is equal to the *default spectrum*.

The solution of the unfolding problem also requires an estimate of the unknown measure error, by means, for instance, of some assumption about the counting statistics. In particular, it is assumed that the measure errors are normally distributed with zero mean and standard deviation σ_k. Thus, the *chi-square* per degree of freedom χ^2 can be introduced as follows:

$$\chi^2 = \frac{1}{n}\sum_{k=1}^{m}\frac{\varepsilon_k^2}{\sigma_k^2} \tag{1.3}$$

It is worth noting that χ^2 is equal to one if $\varepsilon_k^2 \approx \sigma_k^2$.

With this method, a solution of the unfolding problem can be found by maximizing the *Cross Entropy S* with constraints given by Eq. (1.1) and Eq. (1.3). This can be done by using the Lagrange multipliers equation

$$\begin{aligned} L(f_i,\varepsilon_k,\lambda_k,\mu) = &-\sum_{i=1}^{n}\left[f_i \ln\left(\frac{f_i}{f_i^{DEF}}\right) + \left(f_i^{DEF}-f_i\right)\right] \\ &-\sum_{k=1}^{m}\lambda_k\left[\sum_{i=1}^{n}R_{i,k}f_i - I_k - \varepsilon_k\right] \\ &-\mu\left[\sum_{k=1}^{m}\frac{\varepsilon_k^2}{\sigma_k^2}-\chi^2\right] \end{aligned} \tag{1.4}$$

where λ_k and μ are $(m+1)$ Lagrange Multipliers.

The maximum entropy method has been implemented in the code MAXED (Reginatto and Godhagen, 1999), and represents the state-of-the-art for unfolding of neutron spectra. In this work the code MAXED is applied for the first time in X-ray spectrometry. MAXED solves the unfolding by optimizing the maximum entropy Lagrange multipliers equation through a simulated annealing algorithm (Reginatto and Godhagen, 1999). MAXED and GRAVEL (an alternative unfolding code, Matzke, 2002) are contained in the UMG (Unfolding with MAXED and GRAVEL) package version 3.3, released in March 2003, developed by the Physikalisch Technische Bundesanstalt and distributed by NEA Data Bank (Reginatto, 2003). MAXED needs as input parameters the measured spectrum with its variance, the computed DRF, the estimate of the solution called *default spectrum* and the desired precision of the solution expressed through the *chi-square*. It is worth noting that incorrect values of *chi-square* lead to meaningless results (or no results at all). To prevent this problem, the inverse problem solved with GRAVEL provides a suitable estimate of the reachable minimum of the *chi-square* per degree of freedom.

In this article, a new strategy is presented which allows the solution of the unfolding problem in X-ray spectrometry by using a suitable combination of MAXED and GRAVEL. It is implemented within the new code UMESTRAT (Unfolding Maximum Entropy STRATegy), which applies this strategy in a semi-automatic mode.

2. UMESTRAT strategy

UMESTRAT is a graphical tool developed at the University of Bologna to solve the unfolding X-ray spectrometry problem by means of a suitable combination of the codes MAXED and GRAVEL. Given a vector representing the measurement and the DRF matrix, the code computes and plots the unfolded spectrum solution as a function of the energy *E*. Every computation consists of six relevant computational steps (Fig. 1) which are interactively activated by the user

1. Tab Input Mode. The user can decide whether to start a new UMESTRAT project or recover a previous computation.
2. Tab Measured Spectrum. The user loads the measurement data. The input file can be either in the ".phs" format (the file formats used by MAXED and GRAVEL Reginatto, 2003) or ".mca" formats (multichannel analyzer output).
3. Tab Detector Response. The user loads the DRF matrix in the ".rsp" format (file format of MAXED and GRAVEL Reginatto, 2003).
4. Tab Chi Square. GRAVEL is used to compute the initial value of the *chi-square* per degree of freedom in order to ensure the later convergence of MAXED. The unfolding procedure is solved using a positive unitary distribution as an initial *default spectrum* and, as a built-in stop condition, a unitary *chi-square* per degree of freedom. The maximum number of iterations can

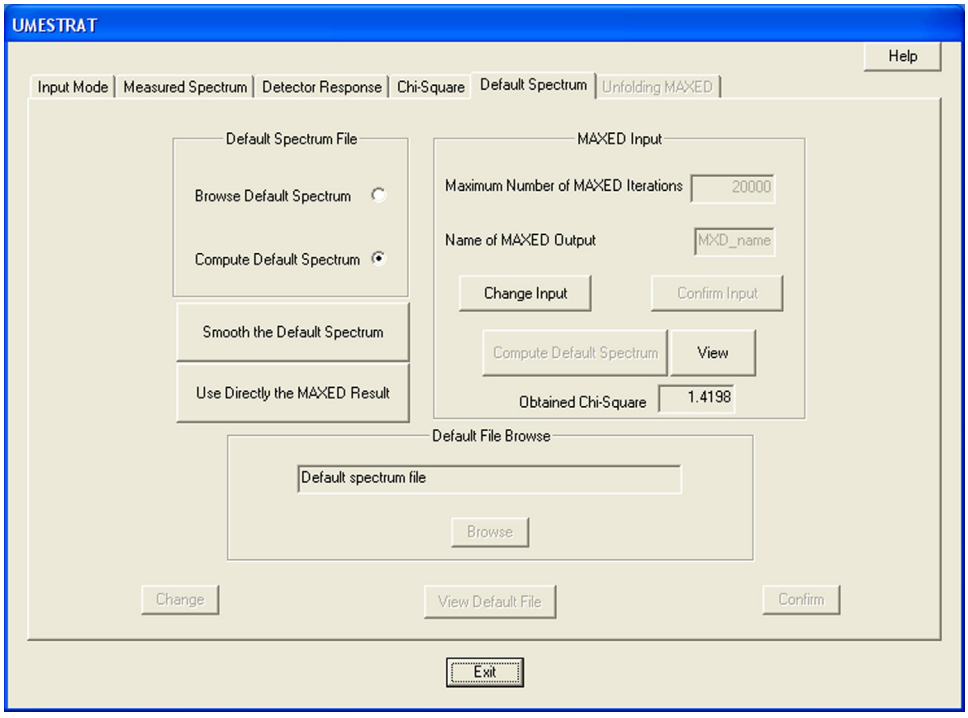

Fig. 1. Screenshot of the code UMESTRAT. The 6 tabs corresponds to the relevant computational steps. Every tab can be accessed after completion of the previous tabs.