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Tensor decompositions for the analysis of atomic resolution electron energy loss spectra

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ABSTRACT

A selection of tensor decomposition techniques is presented for the detection of weak signals in electron energy loss spectroscopy (EELS) data. The focus of the analysis lies on the correct representation of the simulated spatial structure. An analysis scheme for EEL spectra combining two-dimensional and n-way decomposition methods is proposed. In particular, the performance of robust principal component analysis (ROBPCA), Tucker Decompositions using orthogonality constraints (Multilinear Singular Value Decomposition (MLSVD)) and Tucker decomposition without imposed constraints, canonical polyadic decomposition (CPD) and block term decompositions (BTD) on synthetic as well as experimental data is examined.

1. Introduction

Electron Energy Loss Spectroscopy (EELS) is a method which found its way into the standard repertoire of electron microscopy techniques due to its capability to analyze samples simultaneously structurally and chemically on length scales only limited by the microscope's resolution. In principle, this puts the possible resolution of EELS well into the subnanometric regime for modern instruments [1,2]. However, if taken to the extreme of atomic resolution, EEL spectra often suffer from a low signal to noise (S/N) ratio which may render the correct mapping of weak signals, e.g., subtle changes in the fine-structure of an elemental edge due to different chemical environments, difficult.

There are several parameters determining the possible S/N ratio of EEL spectra. For example, an increased beam brightness, dwell time on the respective pixel or increased collection angle all lead to an enhanced S/N ratio. When considering a given setup, the signal to noise ratio of EEL spectra is limited by the stability of the sample with respect to beam damage. This puts a hard limit on the achievable S/N ratio and thereby on the capability to analyze the sample.

Complementary to instrumental efforts to improve the data quality, a statistical data treatment may lead to an improved understanding of the underlying physical information, either by removing the noise (denoising) or by identifying physically meaningful underlying source components of which the measured signal is composed (blind source separation). As the stability of microscopes is steadily increasing, it can be expected that the sizes of EELS data tensors will become increasingly large clearing the ground for a powerful statistical analysis.

Among the most common data processing methods is principal component analysis (PCA) which has been introduced for the analysis of EEL spectra and microscopic image analysis in [3] and has since been used for denoising (e.g., [4]) and physical analysis or EEL spectra, either by direct interpretation of the principal components (e.g., [5]) or by posterior physically motivated rotation of the principal components [6]. As is already commonly known and we demonstrate below, PCA performs poorly in the presence of outliers (e.g., [7,8]). We suggest to apply robust principal component analysis (ROBPCA) as suitable denoising method being robust against outliers and otherwise grossly corrupted spectra in the data set. Among the multitude of available ROBPCA algorithms (for a review see [9]) we focus our analysis on the algorithm of Hubert et al. [10].

Lichtert et al. [11] pointed out that PCA may lead to artifacts in the localization of the denoised physical components if the dimension of the data is not chosen with care. Although we did not observe such types of artifacts in practice, it highlights the necessity of obtaining a clear, correct localization of the source components. A possible road to cleaner coefficient maps lies in the application of tensor decompositions. When going from two-dimensional decomposition methods to a three-dimensional analysis, also the spatial correlations of the EELS data tensors can be considered for an optimal denoising. Especially in the case of atomic resolution EELS data, the data tensor can be often embedded in a much lower dimensional space along the spatial modes. These modes often have a low rank and can be well represented by only few components.

This idea can be grasped by considering a hypothetical EELS or EDX data tensor with a scan area of 50×50 pixels and 2000 energy

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channels. Typically, this tensor would be matricized to a 2500×2000 matrix so that PCA (or any other 2D extraction method) finds the spectral signatures of the source components. However, one can equivalently matricize the tensor to a 100,000×50 matrix. A 2D decomposition method now yields the spatial signatures of the source positions. Loosely speaking, tensor decompositions perform both decompositions simultaneously, so that information about source components as well as positions can be used for the denoising or analysis of the data tensor.

For a more complete overview of available tensor decomposition techniques than those included in the current paper and details on different algorithms, we refer the reader to [12–14] and references therein. Here, the discussion will be focused on Tucker decompositions [15] using either no constraints or orthogonality constraints on all modes, a technique also known as *multilinear singular value decomposition* (MLSVD, or *higher order singular value decomposition* HOSVD) [16], canonical polyadic decomposition (CPD, also *parallel factor analysis* PARAFAC) [17] and Block term decomposition (BTD) [18]. Strengths and shortcomings of these methods when attempting to map a weak component in noisy synthetic data are discussed.

Throughout the article we use small case symbols to indicate scalar and vectorial quantities, bold upper case symbols to indicate matrices and calligraphic upper case symbols to denote tensorial quantities. Following the terminology of the tensor decomposition field of research, the two spatial and single energy dimension of the tensor will be referred to as spatial and energy *mode*, respectively, because the term *dimension* is used to describe the number of modes of the tensor or also the tensor's rank along a specific mode.

2. Creation of synthetic EEL spectra

The model for the synthetic data can be described as

$$\mathcal{X} = Background + Edges + Noise + Spikes$$

The background was modeled by a single power law $B = a \cdot E^{-b}$. The parameter b was gradually changing over the spatial structure of the data set, what resembles the behavior of the background variation in real EELS data, a was kept constant at all times.

The edge signal was composed of three components modeled by two Gaussian peaks with a double step function (two hyperbolic tangens) underneath. The third component was chosen to be an additional fine-structure which was added on top of the second component's edge. By construction the components are not orthogonal to each other, what resembles experimental data sets. In particular, the correlation coefficient of a fine-structure like components (comp. 3) to its edge structure (comp. 2) is 0.26 clearly indicating the non-orthogonality of the two components. These two components may resemble differences of the same elemental species in different chemical environments found in EELS data.

Lastly, noise and outlying spikes were modeled using a Poisson distribution and the Matlab 2014b *salt* & *pepper* noise from which the additive Gaussian noise was subtracted.

The spatial structure of the three source components, which was chosen to be non-negative and strongly overlapping, as well as their spectral signatures are displayed in Fig. 1. The model mimics an atomic resolution data set, where the maps of the three components are strongly overlapping. Of particular interest is the single atom of the third component which is the sample's weakest feature. Its correct retrieval may be taken as indicator for the performance of the analysis method. The size of the synthetic data tensor was $100 \times 100 \times 1000$ leading to a lattice spacing of 10 pixels, what is comparable to commonly achieved magnifications in atomic resolution EELS experiments. In total, three data tensors with identical source components were created.

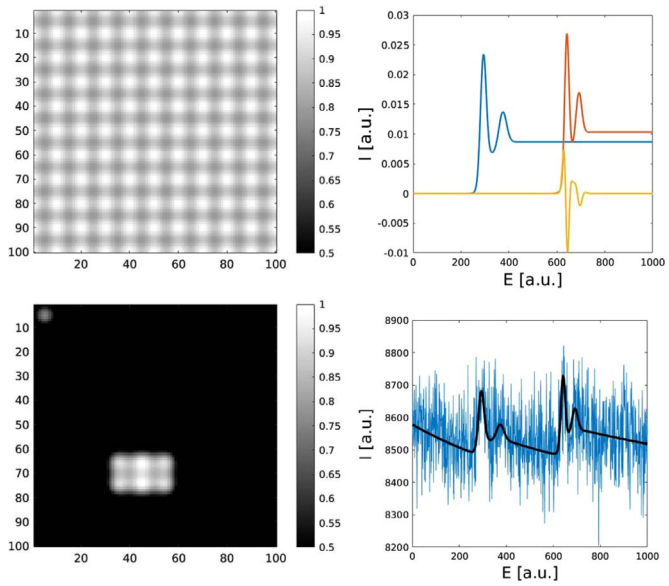

Fig. 1. *Top left*: Spatial structures of source components 1, the structure of component 2 corresponds to that of component 1, where positions of minima and maxima have been swapped. *Bottom left*: Spatial structures of source component 3. The scale bar in both aforementioned images ranges from 0.5 (black) to 1 (white). *Top right*: Spectral signatures of the three source components. *Bottom right*: Example for a synthetic spectrum. Superposed is the noise-free spectrum where the two simulated edge regions can be identified.

3. Procedure for data compression

When faced with the problem of analyzing atomic resolution EELS data the first necessary step is to denoise the data before subjecting them to any further analysis. This goal can be achieved either by local filtering (such as median or gaussian filters) or by decomposing the data and reconstructing them using only a limited number of components - by reducing the dimension of the data. Since local filters often blur the spatial structure or the spectral signatures, it is desirable to achieve the denoising using the latter approach. In a typical experiment, several data tensors are measured. Not all of them may contain an interesting structure, while still being describable by the same source components as the physically interesting data tensor. For a better denoising result, it is therefore advisable to treat all data tensors simultaneously for the identification of the physically relevant signal subspace. Since the spatial structures may vary strongly between different data tensors and the size of their modes may not match, this simultaneous treatment can often not be done using tensor decompositions. We propose to perform the simultaneous denoising of the energy mode instead using 2D decomposition methods such as (ROB) PCA. In a second denoising step, tensor decompositions can be applied to the data tensor of interest to further denoise the spatial structure of the data tensor.

Since both techniques are robust against stochastic noise, lead to a unique solution and allow for an easy identification of the signal subspace, we suggest ROBPCA and MLSVD for the denoising of EELS (or EDX) data tensors due to their favorable properties discussed below.

3.1. Robust principal component analysis

For this first step of the analysis, the spatial modes of the three synthetic data tensors were unfolded, matricized. Since spatial structures are not considered in ROBPCA, it does not matter that they are identical in that regard. The inclusion of sharp spikes creates a number of strongly outlying data points which will disturb a classical PCA decomposition. Like classical PCA, ROBPCA of the joint data matrix **X**