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EFTEM spectrum imaging at high-energy resolution

Bernhard Schaffer*, Gerald Kothleitner, Werner Grogger

Research Institute for Electron Microscopy, Graz University of Technology, Steyrergasse 17, A-8010 Graz, Austria

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Abstract

This paper deals with the application of high-energy resolution EFTEM image series and the corrections needed for reliable data interpretation. The detail of spectral information gained from an image series is largely determined by the intrinsic energy resolution. In this work we show that energy resolution values of as low as 0.8 eV in spectra extracted from EFTEM image series can be obtained with a small energy-selecting slit. At this resolution level aberrations of the energy filter, in particular the non-isochromaticity, can no longer be neglected. We show that the four most prominent factors for EFTEM image series data correction—spatial drift, non-isochromaticity, energy drift and image distortion—must not be treated independently but have to be corrected in unison. We present an efficient algorithm for this correction, and demonstrate the applied correction for the case of a GaN/AlN multilayer sample.

Keywords: EFTEM spectrum imaging; Spectral aberration; Sample drift; Energy drift; Non-isochromaticity; Data correction

1. Introduction

Energy filtering transmission electron microscopy (EFTEM) is nowadays a well-established method in many areas of materials research [1]. Image contrast and resolution in bright field images can be enhanced by using only elastically scattered electrons (zero-loss filtering) [2,3], and electrons in the low energy-loss region often show distinct material contrast (contrast tuning) [4]. Furthermore, elemental information can be quickly mapped at high spatial resolution by combination of a few EFTEM images (elemental maps, jump ratio images) [5].

However, the 'full' spectral information can only be reconstructed from a larger series of EFTEM images taken with a sufficiently small energy-selecting slit, avoiding large energy gaps between the images. This method is known as 'Electron Spectroscopic Imaging' (ESI) [6], 'Image Spectroscopy' [7,8], 'Imaging-Spectrum' [9] and 'EFTEM Spectrum Imaging' (EFTEM SI) [10]. The full 3-dimension dataset gained by the method contains an electron energy-loss (EEL) spectrum for each point of the sampled. This opens up the large range of data analysis methods originally developed for EELS spectroscopy, e.g. plural scattering deconvolution, accurate background subtraction, quantification by multiple least-squares (MLS) fitting, separation of overlapping edges, more accurate sample thickness determination, automated elemental identification, and more. As this information is available for all image pixels, mapping of various associated physical and chemical information becomes possible as shown by numerous authors [7,8,11,12]. In comparison to the alternative method of EELS SI, which uses a focused scanning beam for sequential acquisition of the spectra (STEM EELS SI) [13], EFTEM SI offers high spatial resolution over a larger field of view (many pixels along spatial axes) at comparably short acquisition times. It is therefore often the preferred method when the spatial distributions of spectral features (e.g. ionization edges, plasmon peaks, etc.) need to be mapped over larger sample areas. However, STEM EELS SI offers better energy resolution and collects information over larger energy-loss ranges in one step (many pixels along energy axis). It is therefore most commonly used to investigate subtle spectral changes in small sample areas [14,15].

The aim of this work was to further improve the method of EFTEM SI with respect to energy resolution. The intrinsic energy resolution of an EFTEM SI is a

*Corresponding author. Tel.: +43 316 873 8330; fax: +43 316 811 596.
E-mail address: bernhard.schaffer@felmi-zfe.at (B. Schaffer).

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convolution of the system's total energy resolution (the sum of the energy width of the electron beam, the high tension stability, the spectrometer energy resolution, etc.) with the energy selecting slit width. Considering a total system's energy resolution of 1 eV or better, the energy resolution of the series is then largely determined by the slit width, which in most cases lies in the range of a few to some tens of eV. Some imaging filters allow slit widths smaller than 1 eV to be used so that the energy resolution of the EFTEM SI is no longer limited by the slit and then becomes comparable to the energy resolution of EELS spectra. With the higher energy resolution, additional EELS analysis can be performed and used for mapping, e.g. chemical shift maps, plasmon peak position maps, dielectric constant maps by Kramers–Kronig analysis, etc. Fig. 1 shows another benefit of having better EFTEM energy resolution. In this picture four plasmon ratio maps are compared. Such maps were produced by dividing two EFTEM images acquired at the energy-losses of the GaN and AlN plasmon peaks, respectively. The first three maps (Fig. 1A–C) were produced from datasets acquired with different energy selecting slit widths of 10, 5 and 1 eV, respectively. The datasets were fully corrected for the combined effect of spatial drift, energy drift and non-isochromaticity (NIC), using the novel algorithms described in this work. The fourth map was produced from the same data as the third map, but without data correction except for spatial drift. All images are displayed with the same contrast limits. It can be clearly seen that for best image contrast a small slit width (1 eV) is superior to larger slit widths (5 eV, 10 eV). However, at energy resolutions in the range of the spectrometer aberrations, their influence can no longer be neglected as can be seen from the uncorrected data (Fig. 1D).

This work focuses on the correction of high-energy resolution EFTEM SI data, which becomes essential for reliable data analysis and interpretation. To our best knowledge, it presents the first algorithm, which corrects the combined effect of spatial drift, energy drift and NIC.

2. Data correction

The information contained in an image series can be best visualized in 3-dimensional information space, where x and y are spatial coordinates of the sample and z is the EEL axis as suggested by Jeanguillaume et al. [16]. An EFTEM SI is a discrete subset of this information space as shown in Fig. 2A. Any data point of the EFTEM SI contains the integrated intensity of a small subset of the information space centered at given spatial coordinates and energy-loss with dimensions according to the spatial and energy resolution of the EFTEM SI. In analogy to 'pixel', which is the commonly known acronym for 'picture element', such a data point in a discrete 3-dimensional data set is called 'voxel' as acronym for 'volume picture element'.

Ideally, an EFTEM SI is a cuboid built as a stack of horizontal slices (EFTEM images). However, the real shape of a measured EFTEM SI in information space is more or less deformed due to system instabilities and spectrometer lens aberrations. Nevertheless, these deformations are often neglected and the EFTEM SI is treated as an orthogonal block leading to serious errors if used for further data evaluation without corrections. The most influential factors are shown in Figs. 2C–F, leading to a combined deformation shown in Fig. 2B. The deformed EFTEM SI is falsely interpreted as the cuboid shown by the dashed line according to the nominal values of the EFTEM SI acquisition.

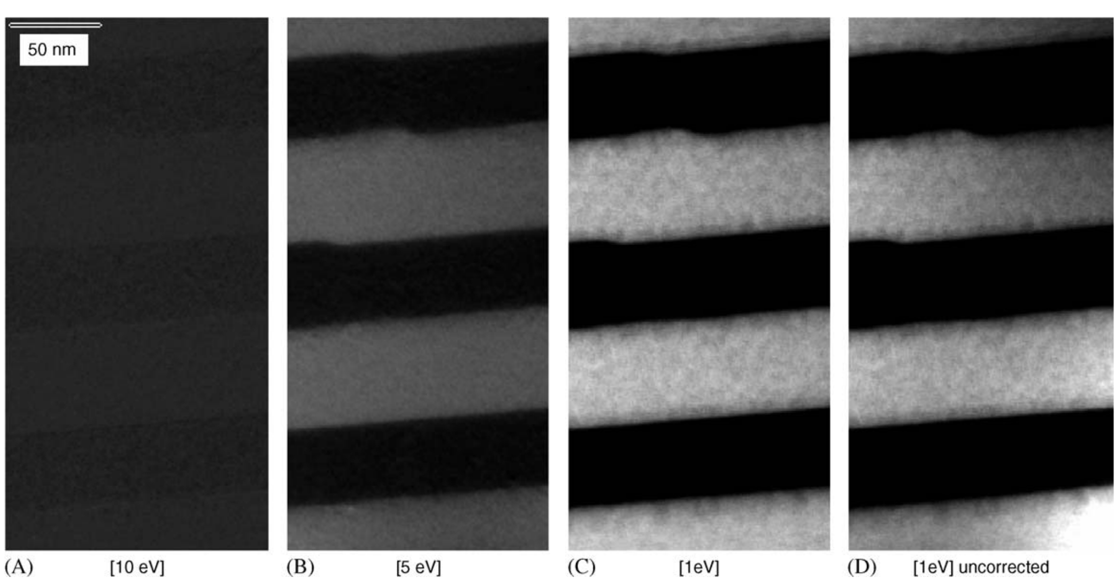

Fig. 1. Plasmon ratio maps (20.8 eV image dived by a 19.5 eV image) of a GaN/AlN stack acquired with different energy selecting slit widths (A: 10 eV; B: 5 eV; C and D: 1 eV). The maps are displayed with the same contrast settings and were produced from fully corrected data except image D, where no non-isochromaticity correction was performed.