StatSTEM: An efficient program for accurate and precise model-based quantification of atomic resolution electron microscopy images

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StatSTEM: An efficient program for accurate and precise model-based quantification of atomic resolution electron microscopy images

A De Backer[1], K H W van den Bos[1], W Van den Broek[2], J Sijbers[3], and S Van Aert [1]

[1] Electron Microscopy for Materials Science (EMAT), University of Antwerp, Groenenborgerlaan 171, 2020 Antwerp, Belgium
[2] Ag Strukturforschung/Elektronenmikroskopie, Institut für Physik, Humboldt-Universität zu Berlin, Newtonstraße, 12489 Berlin, Germany
[3] imec-Vision Lab, University of Antwerp, Universiteitsplein 1, 2610 Antwerp, Belgium

E-mail: annick.debacker@uantwerpen.be

Abstract. An efficient model-based estimation algorithm is introduced in order to quantify the atomic column positions and intensities from atomic resolution (scanning) transmission electron microscopy ((S)TEM) images. This algorithm uses the least squares estimator on image segments containing individual columns fully accounting for the overlap between neighbouring columns, enabling the analysis of a large field of view. For this algorithm, the accuracy and precision with which measurements for the atomic column positions and scattering cross-sections from annular dark field (ADF) STEM images can be estimated, is investigated. The highest attainable precision is reached even for low dose images. Furthermore, advantages of the model-based approach taking into account overlap between neighbouring columns are highlighted. To provide end-users this well-established quantification method, a user friendly program, StatSTEM, is developed which is freely available under a GNU public license.

1. Introduction

Nowadays, the field of nanotechnology requires more and more quantitative characterisations of nanomaterials. This includes for example locating atomic column positions with picometre precision [1], counting the number of atoms with single atom sensitivity [2–4] and precise chemical composition determination [5, 6]. In order to facilitate these needs, we developed a user-friendly software package called StatSTEM [7]. This program allows to apply advanced image quantification methods , such as atom-counting, measuring atomic column positions, and strain-mapping, to atomic resolution electron microscopy images by using an efficient model-based fitting algorithm. This software package is freely available under the GNU public license and can be found at the website [8]. A screen shot of the program is given in Fig. 1.

2. Model-based parameter estimation and the efficient model estimation algorithm

For atomic resolution (scanning) transmission electron microscopy ((S)TEM) images, statistical parameter estimation theory provides an excellent tool to quantitatively extract unknown structure parameters [9, 10]. In these atomically resolved images, the intensity is peaked at the

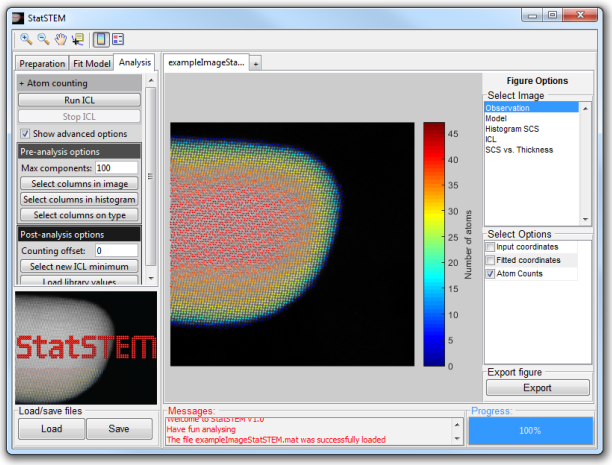

Figure 1. A screen shot of the StatSTEM program, showing atom-counting results on an experimental image of Au nanorod.

atomic column position and can be modelled as a Gaussian function [11, 12]. The expectation of the intensity of pixel (k, l) at position (x_k, y_l) can then be described by an expectation model $f_{kl}(\boldsymbol{\theta})$ with $\boldsymbol{\theta}$ the vector of unknown structure parameters, as:

$$f_{kl}(\boldsymbol{\theta}) = \zeta + \sum_{i=1}^{I} \sum_{m_i}^{M_i} \eta_{m_i} \exp\left(-\frac{\left(x_k - \beta_{x_{m_i}}\right)^2 + \left(y_l - \beta_{y_{m_i}}\right)^2}{2\rho_i^2} \right) \tag{1}$$

with ζ a constant background, ρ_i the column dependent width of the Gaussian peak, η_{m_i} the column intensity of the m_ith Gaussian peak, $\beta_{x_{m_i}}$ and $\beta_{y_{m_i}}$ the x- and y-coordinate of the m_ith atomic column respectively. The index i refers to atomic columns of the same atom type with I different types and the index m_i refers to the mth column of type i with M_i the number of columns of type i. The model includes a constant background which can account for the black level of the detector or for a constant contribution of a sample support. The indices in the summation of Eq. (1) can be simplified in the case of a mono-type crystalline nanostructure since then only one column type is present.
The parametric model of Eq. (1) is fitted to the experimental data using the uniformly weighted least squares criterion quantifying the similarity between the experimental images and the model. A direct implementation of the least squares estimator, where all parameters are simultaneously estimated, is computationally very intensive and can only be applied when dealing with a limited number of projected atomic columns. Therefore, we proposed a new, efficient algorithm [7]. This new algorithm divides the image into smaller segments containing individual columns without ignoring the overlap of image intensities between neighbouring columns. This overlap is estimated in an iterative procedure by subtracting in each iteration step the estimated contributions of neighbouring columns from the small segment of the single atomic column.

3. Benefits of a model-based approach

In order to illustrate the benefits of this new method in a statistical manner, the accuracy and precision are evaluated for the estimated atomic column position and the total scattered intensity of an atomic column, i.e. the so-called scattering cross-section which equals the volume, $V = 2\pi\eta_{m_i}\rho_i^2$, under the estimated Gaussian peak and which can be used to quantify the chemical composition and the number of atoms. This evaluation is performed as a function of thickness for the central atomic column of a simulated ADF STEM image of Pt along the [100] zone axis

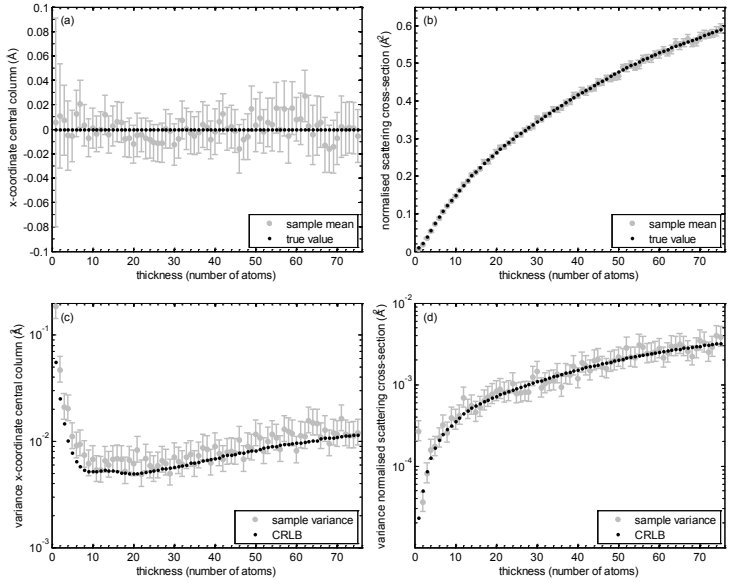

Figure 2. Accuracy and precision of the estimated position coordinate and scattering cross-section as a function of thickness in a Pt crystal for an incoming electron dose of $5 \cdot 10^2 \mathrm{e}^- \mathrm{Å}^{-2}$. (a,b) The true values and the sample mean together with the 95% confidence intervals. (c,d) The highest attainable precision, the CRLB, and the sample variance together with the 95% confidence intervals.

orientation. Using 100 noise realisations for an incident electron dose of $5 \cdot 10^2 \mathrm{e}^- \mathrm{Å}^{-2}$, the sample mean is compared with the true value in order to evaluate the accuracy. In order to evaluate the precision, the sample variance is compared with the theoretical lower bound on the variance, the so-called Cramér-Rao lower bound (CRLB). It is shown that even for these low electron dose images structure parameters are accurately measured with the highest attainable precision (Fig. 2).

Furthermore, the advantages of our model-based approach are illustrated as compared to the computationally less expensive approach where the overlap between neighbouring columns is neglected. This is first shown by estimating the spacing between two neighbouring Pt atomic columns of 5 and 3 atoms thick as a function of the distance (Fig. 3a) when taking overlap into account and when neglecting it (Fig. 3b). Secondly, a comparison between the estimated volumes and the integrated intensities from a Voronoi cell reveals that the estimated volumes give more accurate results for predicting the scattering cross-section (Fig. 3c). Therefore, this comparison further highlights the advantages of our model-based approach.

4. Conclusions

In conclusion, a new efficient algorithm is developed enabling a quantitative analysis of (S)TEM images having a large field of view. As this algorithm still accounts for the overlap of image intensities between neighbouring columns, it provides accurate measurements of structure parameters with the highest attainable precision. To provide end-users this powerful quantification method, a user-friendly software package is available.

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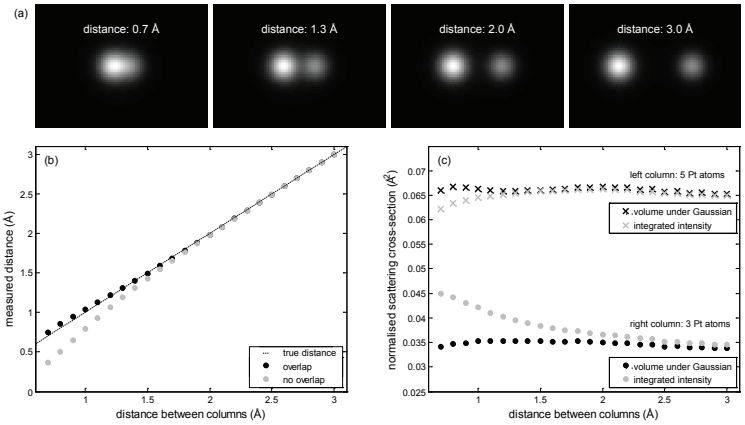

Figure 3. Influence of a neighbouring column on the estimated parameters. (a) Examples of simulated images of a 5 and 3 Pt thick column with a varying distance between the columns. (b) The estimated distance when taking the overlap of neighbouring columns into account and when neglecting it. (c) The estimated scattering cross-sections obtained from the volumes under the fitted Gaussian functions and from integrating intensities from Voronoi cells.

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