Ultramicroscopy 187 (2018) 84–92

Contents lists available at ScienceDirect

Ultramicroscopy

journal homepage: www.elsevier.com/locate/ultramic

Thickness dependence of scattering cross-sections in quantitative scanning transmission electron microscopy

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ARTICLE INFO

Article history:
Received 27 June 2017
Revised 16 January 2018
Accepted 17 January 2018
Available online 31 January 2018

Keywords:
ADF STEM
Electron scattering
Quantitative STEM
Image simulation

ABSTRACT

In quantitative scanning transmission electron microscopy (STEM), scattering cross-sections have been shown to be very sensitive to the number of atoms in a column and its composition. They correspond to the integrated intensity over the atomic column and they outperform other measures. As compared to atomic column peak intensities, which saturate at a given thickness, scattering cross-sections increase monotonically. A study of the electron wave propagation is presented to explain the sensitivity of the scattering cross-sections. Based on the multislice algorithm, we analyse the wave propagation inside the crystal and its link to the scattered signal for the different probe positions contained in the scattering cross-section for detector collection in the low-, middle- and high-angle regimes. The influence to the signal from scattering of neighbouring columns is also discussed.

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1. Introduction

Quantitative scanning transmission electron microscopy (STEM) has become a widely used technique to retrieve structural information from a material under study. It generally refers to the analysis of images obtained using a scanning transmission electron microscope in which the electrons are usually collected using an annular detector, although detectors with different configurations are also becoming more widely available [1–5]. The choice of inner and outer collection angles of an annular detector allows one to collect electrons scattered over a specific angular range resulting in specific structural information of the sample. Electrons that scatter at high angles are related to the atomic number Z of the atoms they interact with, allowing so-called Z-contrast imaging [6,7]. Other collection angle regimes enable imaging of light elements or strained samples [8–10]. In the annular dark field (ADF) regime, the signal is considered predominantly incoherent [11,12], which facilitates the interpretation of the images due to the lack of contrast reversals. In this collection angle regime quantitative methods have been developed to analyse images. The proposed quantitative methodologies usually follow either an image simulation-based or a statistics-based approach. Image simulation-based methods depend on the direct comparison of experimental images with simulations [13–19]. The experimental images need to be normalised with respect to the incident beam [14,16,20]. In addition, image simulations need to be performed using models that describe the electron-sample interaction as accurately as possible [21–27]. Furthermore, the experimental conditions [28] as well as the detector's response [19,20,29–32] need to be known up to the measurable limits. On the other hand, statistics-based methods consider a statistical parameter estimation framework to extract quantitative information from STEM images [33,34]. They make use of parametric models of which the unknown parameters are estimated by fitting the model to the experimental images using a criterion of goodness of fit. The quantitative information is then retrieved from these estimated model parameters and can be directly related to the chemical composition [34,35] or number of atoms in an atomic column [36,37], for example. When combined with tomography techniques, these methods allow the three-dimensional reconstruction of a material at the atomic level [38–40] without the need of image simulations. The tools of statistical experimental design can be used to find the optimal microscope settings to retrieve the information of interest [41–44]. It has also been shown that a combination of statistics-based and image simulation-based methods leads to both accurate and precise structure parameters [20,35,36,43,45,46].

Different measures have been used to quantify STEM images including peak intensities at the atomic column position or contrast variations [7,31], the mean intensity of the material's unit cell [10,16,17,30], the volume under a Gaussian function fitted at the atomic column position [34–37] or the pixel integrated

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https://doi.org/10.1016/j.ultramic.2018.01.005
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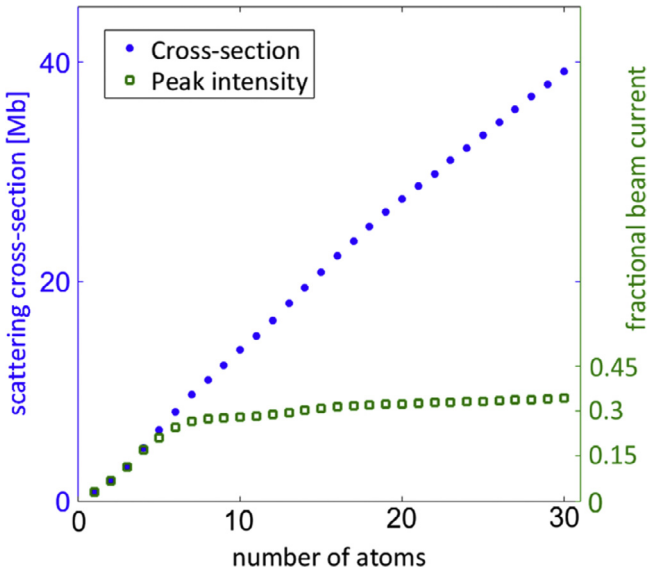

Fig. 1. Scattering cross-section (left blue vertical-axis) and peak intensity (right green vertical-axis) as a function of sample thickness for Pt in [110] zone axis. ADF STEM detector collection angles ranging from 63 to 200 mrad for a 21 mrad probe convergence angle (See Table 1). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 1
Frozen lattice simulations settings

Acceleration voltage HT	300 kV
Defocus C_1	0 nm
Spherical aberration C_3	−142 nm
Convergence semi-angle α	21 mrad /31 mrad
Spatial incoherence of source FWHM	0.7 Å
LAADF inner collection angle	21 mrad /31 mrad
MAADF inner collection angle	42 mrad /62 mrad
HAADF inner collection angle	63 mrad /93 mrad
LAADF/MAADF/HAADF outer collection angle	200 mrad
Detector's response	homogeneous (ideal)
Zone axis	[110]
Supercell periodicity	9 × 9 unit cells
Supercell size	24.97 × 35.31 Å^2
Maximum specimen thickness	8.32 nm
Number of phonon configurations	20
Pixel size of simulated image	0.1394 Å
Pixel size to sample atomic potential	0.0293 Å

scattering cross-sections [18,20,47]. The latter two have been found to be equivalent and are referred to as the so-called scattering cross-section of an atomic column, which corresponds to the total amount of scattered electrons by a single atomic column. A detailed derivation of this measure can be found in [47].

De Backer et al. [44] showed that in order to precisely count the number of atoms, the scattering cross-sections perform as well as when comparing images and simulations on a pixel by pixel basis. Furthermore, peak intensities, which correspond to the maximum intensity recorded at the atomic column position, have been found to provide less reliable results. The scattering cross-section has also been found to be robust to probe parameters in contrast to peak intensities [47,48]. Previous experimental work has quantified Pt systems using scattering cross-sections [18,20,43,46]. Therefore, we present an in-depth simulation study of a Pt system to investigate the sensitivity of scattering cross-sections with respect to thickness. In Fig. 1 the increase of scattering cross-section (left blue vertical-axis) and peak intensity (right green vertical-axis) is shown as a function of sample thickness for a Pt crystal viewed along the [110] zone axis under high angle annular dark field (HAADF) conditions for a 21 mrad probe convergence angle (See Table 1). Scattering cross-section axis units are Megabarn ($1Mb = 10^{-22}m^2$) and for peak intensity, the axis is in fractional beam current. From this plot, it is clear that peak intensities saturate around $\approx$ 6 atoms, whereas the scattering cross-sections keep increasing with increasing thickness. Therefore, when trying to distinguish between e.g. 10 and 11 atoms, the peak intensity measure is not adequate since there is practically no variation between the corresponding intensity values. On the other hand, when using the scattering cross-section values, a clear difference is observed.

In this work, we explain the monotonically increase of scattering cross-sections for the number of atoms in a sample as shown in Fig. 1. For this, we analyse how the probe propagates through the crystal and how this process contributes to the peak intensity and to the scattering cross-section values. Consequently, a set of image simulations describing the wavefunction propagation through the crystal for different probe positions has been carried out by using the multislice algorithm [49,50]. With the information these calculations provide, we are able to infer the origin of the detected signal and its contribution to the scattering cross-section value.

2. Background and methods

In order to characterise the atomic columns in 3D from a STEM image, one needs to use the image intensities that originate

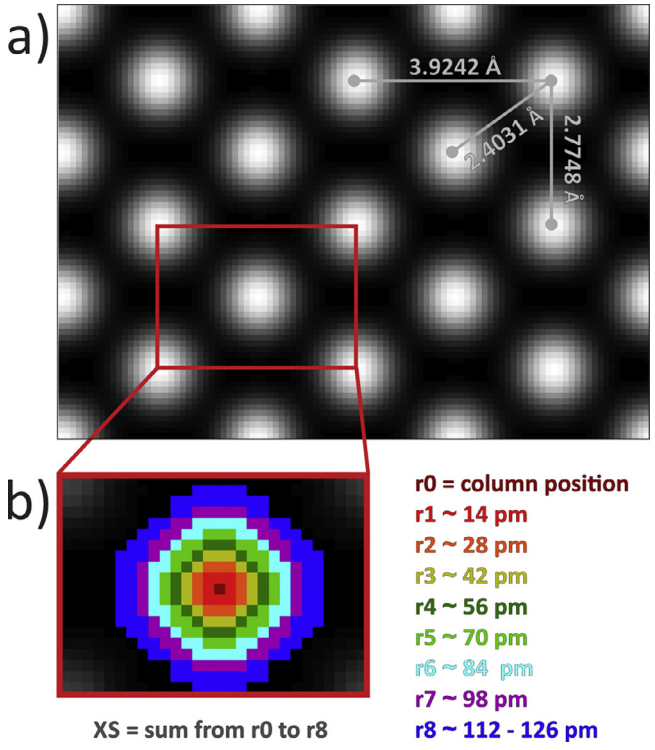

Fig. 2. a) Simulated image of Pt in [110] zone axis using simulation settings described in Table 1 for the HAADF case using a 21 mrad probe convergence angle. Depicted distances show the separation between columns for this material. b) Colour-edited simulated image of Pt and colour labels with their corresponding distances from the atomic column position. The label **XS** refers to the scattering cross-section, which is the summation over all radial probe positions. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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