doi:10.1017/S1431927620013951

Microsc. Microanal. 26 (Suppl 2), 2020

Quantitative Simulation of Four-dimensional STEM Datasets

Andreas Beyer[1], Saleh Firoozabadi[1], Damien Heimes[1], Pirmin Kükelhan[1], Tim Grieb[2], Florian Krause[2], Marco Schowalter[2], Hoel Laurent Robert[3], Knut Müller-Caspary[3], Andreas Rosenauer[2] and Kerstin Volz[1]

[1]Faculty of Physics and Materials Sciences Center, Philipps-Universität Marburg, Marburg, Hessen, Germany, [2]Universität Bremen, Germany Institut für Festkörperphysik Department for Electron Microscopy, Bremen, Bremen, Germany, [3]Forschungszentrum Jülich, Jülich, Nordrhein-Westfalen, Germany

Recently, four dimensional scanning transmission electron microscopy (4D STEM) emerged as a promising technique for sample characterization due to the multitude of different signals which can be created from the data sets after acquisition (Ophus, 2019). Synthetic images like annular dark field (ADF), annular bright field (ABF) or center of mass (COM) yield useful information about a sample´s structure, composition or even fields present within the sample.

Especially ABF and COM are generated by electrons which are scattered at rather small angles as compared to HAADF (high angle annular dark field) STEM. This has implications if it comes to complementary image simulation. In the case of HAADF STEM, multi-slice simulations are well established and quantitative agreement with the experiment can be achieved, see e.g. (Beyer et al., 2016). This allows to quantify the local composition and structure of experimental images. However, at low scattering angles involved in 4D STEM there are significant deviations between simulation and experiment (Müller-Caspary et al., 2016).

Here we investigate three potential sources of discrepancy, i.e. the presence of amorphous layers from sample preparation, the neglection of plasmon scattering, and the assumption of uncorrelated phonons. To this end, we use a double aberration corrected JEOL JEM 2200 FS equipped with an in-column energy filter and a pixelated pnCCD camera. This allows for the acquisition of atomically resolved 4D STEM data sets selecting different energy ranges from the electron energy loss spectrum. As well defined sample, we prepared a lamella of silicon in [010] projection exhibiting discrete thickness steps via focused ion beam (FIB) milling with a JEOL JIB 4601. Final Ar ion milling was carried out with a Fischione NanoMill 1040 to control the thickness of amorphous layers on the surfaces of the specimen.

With the help of state-of-the-art image simulations (Oelerich et al., 2017; Rosenauer & Schowalter, 2008), we elucidate and quantify the individual contributions to the scattering. Simulated and experimental data sets reveal that all three investigated factors, i.e. plasmon scattering, the presence of amorphous layers as well as the correlated movement of the sample atoms, significantly contribute to the low angle signal. By correctly implementing these effects into the image simulations, the fit between simulation and experiment at low scattering angles can be improved significantly.

This contribution summarizes the amendments which have to be made to multi-slice simulations to be able to simulate 4D-STEM data sets quantitatively.

References

Beyer, A., Belz, J., Knaub, N., Jandieri, K. & Volz, K. (2016). Influence of spatial and temporal coherences on atomic resolution high angle annular dark field imaging. *Ultramicroscopy* **169**, 1–10. http://linkinghub.elsevier.com/retrieve/pii/S0304399116300845.

Müller-Caspary, K., Oppermann, O., Grieb, T., Krause, F. F., Rosenauer, A., Schowalter, M., Mehrtens, T., Beyer, A., Volz, K. & Potapov, P. (2016). Materials characterisation by angle-resolved scanning transmission electron microscopy. *Scientific Reports* **6**, 37146. http://www.nature.com/articles/srep37146.
Oelerich, J. O., Duschek, L., Belz, J., Beyer, A., Baranovskii, S. D. & Volz, K. (2017). STEMsalabim: A high-performance computing cluster friendly code for scanning transmission electron microscopy image simulations of thin specimens. *Ultramicroscopy* **177**, 91–96. http://linkinghub.elsevier.com/retrieve/pii/S030439911630300X.
Ophus, C. (2019). Four-Dimensional Scanning Transmission Electron Microscopy (4D-STEM): From Scanning Nanodiffraction to Ptychography and Beyond. *Microscopy and Microanalysis* 1–20. https://www.cambridge.org/core/product/identifier/S1431927619000497/type/journal_article.
Rosenauer, A. & Schowalter, M. (2008). STEMSIM - A New Software Tool for Simulation of STEM. *Microscopy of Semiconducting Materials 2007* **120**, 170–172. http://www.springerlink.com/index/j2344u6684318134.pdf (Accessed December 8, 2011).