Quantitative STEM - From composition to atomic electric fields

Authors: Andreas Rosenauer (1), Knut Müller-Caspary (1), Marco Schowalter (1), Tim Grieb (1), Florian F. Krause (1), Thorsten Mehr tens (1), Armand Béché (2), Johan Verbeeck (2), Josef Zweck (3), Stefan Löfler (4), Peter Schattschneider (4), Marcus Müller (5), Peter Veit (5), Sebastian Metzner (5), Frank Bertram (5), Jürgen Christen (5), Tillmann Schimpke (6), Martin Strassburg (6), Rafal E. Dunin-Borkowski (7), Florian Winkler (7), Martial Duchamp (7)

1. Institut für Festkörperphysik, Universität Bremen, Bremen, GERMANY
2. EMAT, University of Antwerp, Antwerp, BELGIUM
3. Institut für Experimentelle und Angewandte Physik, Universität Regensburg, Regensburg, GERMANY
4. USTEM, Vienna University of Technology, Vienna, AUSTRIA
5. Institute of Experimental Physics, Otto-von-Guericke-University Magdeburg, Magdeburg, GERMANY
6. OSRAM, Opto Semiconductors GmbH, Regensburg, GERMANY
7. ER-C and PGI, Forschungszentrum Jülich, Jülich, GERMANY

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Corresponding email: rosenauer@ifp.uni-bremen.de

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The image intensity in high-angle annular dark field STEM images shows a strong chemical sensitivity. As it is also influenced by specimen thickness, crystal orientation as well as characteristics of illumination and detector, a standard-free quantification of composition requires a comparison with accurate image simulation, for which we use the frozen lattice approach of the STEMsim program taking the non-uniform detector sensitivity into account. The experimental STEM intensity is normalized with respect to the incident electron beam. For the quantification of a STEM image it is subdivided into Voronoi cells in which the intensity is averaged. Analysis of the composition in a ternary semiconductor layer such as In$_x$Ga$_{1-x}$N requires measuring the specimen thickness in regions with known composition by comparison with the simulated STEM intensity. Interpolation of the obtained thickness into the layer with unknown composition yields a map of the specimen thickness. Finally, specimen thickness and STEM intensity are compared with simulations computed as a function of composition resulting in a map of the In-concentration $x$. In alloys containing atoms with different covalent radii (e.g. In and Ga in In$_x$Ga$_{1-x}$N) static atomic displacements occur, which are computed with empirical potentials and included in the simulation. As an application example Fig. 1a shows an array of core-shell nanowires. One single nanowire is depicted in Fig. 1b. The core-shell area marked by a yellow frame is shown in Fig. 1c. Figs. 1d and 1f show high-resolution STEM images of the core-shell regions corresponding to the top and the bottom of a nanowire, respectively. The maps of the measured In-concentration given in Figs. 1e and 1g reveal an increasing thickness of the layer along the growth direction. In the upper part, the layer shows variations of the In-concentration clearly beyond the random-array fluctuations as was shown by a comparison with image simulation.

In the second part of the talk we present results on measurements of atomic electric fields. Differential phase contrast STEM detects the field-induced angular deflection of the electron beam with a segmented ring detector (J. Chapman et al., Ultramicroscopy 3 (1978), 203) assuming that the Ronchigram is homogeneously filled and shifted as a whole in the presence of electromagnetic fields (N. Shibata et al., Nat. Phys. 8 (2012), 611). These assumptions were tested by simulation for 1.3 nm thick GaN. Fig. 2b shows Ronchigrams simulated for 6x6 scan positions within the region marked in Fig. 2a. The dominant effect of the atomic electric field is a complex redistribution of intensity within a Ronchigram. By fundamental quantum mechanical arguments, we take the complex intensity distribution in the Ronchigram into account (K. Müller et al., Nat. Commun. 5 (2014), 5653). The intensity in a certain pixel of the recorded Ronchigram is proportional to the probability that the corresponding momentum is observed. Thus, a center-of-gravity type summation yields the expectation value for the momentum. To relate the electric field in the specimen to the observed momentum transfer, Ehrenfest’s theorem is applied. For thin specimens, the expectation value of the momentum is found to be proportional to the projection of the electric field along the optical axis, convolved with the intensity distribution of the incident STEM probe. We demonstrate the potential of this approach in both simulation and experiment. For the GaN simulation in Fig. 2c we find the electric field depicted in Fig. 2d. Atomic sites appear as sources of the field which has a magnitude of up to 1.5 V/pm. As only the convolution of the true field with the probe intensity can be measured, the field strength decreases in the direct vicinity of atomic sites. In a first experiment, 20x20 Ronchigrams of SrTiO$_3$ with a thickness of 2.5 nm have been recorded on a conventional charge-coupled device (CCD), yielding the electric field in Fig. 2e. We also report on pilot experiments with the ultrafast pnCCD camera (K. Müller et al., Appl. Phys. Lett. 101 (2012), 212110) which was operated at read-out rates of up to 4 kHz. For example, Fig. 2f shows the momentum transfers recorded at a MoS$_2$ mono/bilayer interface, demonstrating that fast detectors are the key for atomic-scale materials analyses at a reasonable field of view.
Fig 1: (a) SEM image of a nanorod array, (b) STEM image of a single nanorod viewed in [11-20] projection, (c) STEM image of the core-shell region marked in (b) showing the InGaN quantum well (QW), (d) high resolution STEM image of the InGaN QW from the upper part and (f) from the bottom part of a nanowire. The In-concentration maps (e) and (g) correspond to (d) and (f), respectively.

Fig 2: Simulation study of GaN. (a) Projected potential, (b) simulated Ronchigrams for scanning a 50pm probe in the dashed region of (a), (c) momentum transfers and (d) electric field for the region in (a). (e) Experimentally measured electric field in SrTiO3. (d,e) Contain the bright field signal as background. (f) Momentum transfers for a mono-/bilayer interface in MoS2 on a 256x256 STEM raster.