Quantitative High-Resolution TEM/STEM and Diffraction

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Composition determination using HAADF-STEM in AlGaN/GaN heterostructures revisited

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In recent years methods for quantification of composition from high-angle annular dark field (HAADF) images have been developed and applied to various semiconductor heterostructures by several groups (e.g. [1,2,3]). Usually the HAADF intensity averaged for each atom column is normalized with respect to a region, where the composition is known. The composition is then determined by comparing the measured intensity ratio with reference measurements or image simulations. Principally the normalization is done to minimize the thickness dependence of the evaluation. However, Rosenauer et al. [3,4] showed that the specimen thickness can be directly determined from the HAADF intensity by normalizing the HAADF intensity to the intensity of the incoming electron beam, that is determined by scanning the electron beam over the detector and taking into account the non-uniform sensitivity of the HAADF detector in the image simulations. For the simulations all kinds of disorder in the crystal need to be taken into account, as disorder strongly influences the HAADF intensity. In a pure and perfect crystal disorder is caused only by thermal vibration of the atoms giving rise to thermal diffuse scattering (TDS). In mixed crystals, where e.g. an atomic species is replaced statistically by another one, the different covalent radii cause static atomic displacements (SADs) around a substitutional atom [1]. Due to the similar covalent radii of Al (0.121(4) nm) and Ga (0.122(3) nm) [5] the lattice parameter of Al(group V) and Ga(group V) binary compounds are similar. Therefore, SADs have been neglected in the publication of Rosenauer et al. [3] on quantification of Al concentration in AlGaN. However, despite the similar covalent radii of Al and Ga the lattice parameter mismatch between AlN and GaN is 2.8%. In this contribution we study the effect of SADs on the composition quantification in AlGaN/GaN heterostructures. Typically SADs are taken into account by relaxing the atom positions of the simulation cell using empirical potentials such as the Stillinger-Weber potential [6]. We parametrized the Stillinger-Weber potential (SWP) to describe lattice parameters, elastic constants as well as the binding energies of AlN and GaN. We performed a test of the applicability of our parametrization to SADs: a series of supercells containing 64 atoms and different Al concentrations were set up and relaxed within density functional theory (DFT) approach as implemented in the VASP code [7]. The same supercells were afterwards relaxed using our parameterization and final positions were compared. Maximum SADs were observed to be in the order of 5-6 pm, whereas the maximum shifts between the final positions from DFT and from the SWP were approximately 1-2 pm, which is very close to the maximum reachable accuracy for the potential. The parametrization was then used for the relaxation of atom positions in the supercells for the image simulations. In this way a reference dataset of the average image intensity as a function of the specimen thickness and the Al concentration was obtained. A second dataset, where SADs have been neglected, still was available from the investigation in Ref. [3]. For the comparison a sample consisting of 6 layers with varying Al concentration was used. The Al concentration of each layer was evaluated as described in Ref. [3] using the two different datasets, respectively. In addition the Al concentrations within each layer were determined from strain state analysis (SSA) of the same images. Fig. 1 shows the measured concentrations as a function of the nominal concentrations as points and stars as a function of the nominal concentration determined from calibration samples using X-ray diffraction. The nominal Al concentration and its error region are displayed as the black line and the grey shaded region, respectively. For concentrations below about 50 % the comparison shows that the neglect of SADs in the image simulations is in agreement with the reference measurements. A discrepancy of the measured Al concentration and the nominal one was found for the datapoint at 58 % Al. For this datapoint a nominal concentration of larger than 58 % was given, because the concentration was determined by an extrapolation of the fluxes. However, taking into account the SADs for the image evaluation improves the accuracy of the measurement.

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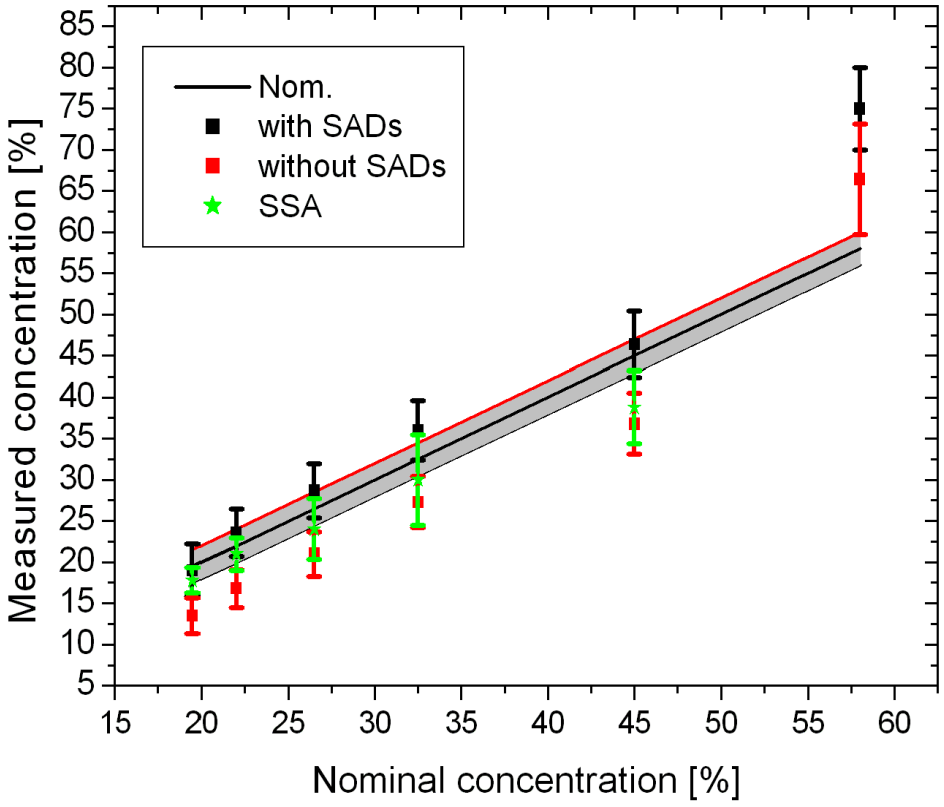

Figure 1. Al concentrations measured from HAADF-STEM image evaluation taking into account SADs (black squares) and neglecting SADs (red squares), nominal concentrations (black line) and concentrations measured from strain state analysis (green stars) as a function of nominal concentration as determined from X-ray diffraction of respective calibration samples.