2D-composition mapping in InGaN without electron beam induced clustering of indium by STEM HAADF Z-contrast imaging

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Abstract. Investigation of composition in InGaN quantum wells and quantum dots by TEM is hampered by formation of electron beam induced agglomeration of indium, which occurs if the specimen is exposed to the electron beam for a few minutes. In this contribution we demonstrate that compositional analysis of InGaN nanostructures is possible without this artifact if STEM Z-contrast imaging is applied instead of parallel beam illumination. The suggested method for composition analysis in InGaN is based on a comparison of intensity normalized with respect to the incident electron beam with simulated image intensity. Simulations are performed with the STEMsim program using the frozen lattice multislice approximation for which static atomic displacements were taken into account.

1. Introduction
In scanning transmission electron microscopy (STEM) using a high angle annular dark field (HAADF) detector, image intensity depends on atomic numbers of scattering atoms, which allows for simple qualitative interpretation of composition distribution. Quantitative measurement of composition is possible if the measured image intensity is compared with accurate simulations computed with the frozen lattice approximation, as shown by Grillo et al. [1]. In reference [1], relative image intensity was compared with simulation and the specimen thickness was obtained by the projection method. These authors also pointed out that taking into account the static atomic displacements that occur if atoms with different covalent radii occupy the same sublattice of a sphalerite type crystal, improves the accuracy of composition determination significantly. Recently, LeBeau and Stemmer suggested use of image intensity normalized with respect to the incident electron beam for quantitative comparison with simulation [2] using external signal amplifiers. Conditions, for which the internal signal amplifiers of a TITAN 80/300 TEM equipped with a Fischione Model 300 HAADF detector can be used, were investigated in reference [3]. In this publication it was also shown that comparison of experimental and simulated image intensities allows measurement of specimen thickness in GaN and evaluation of composition in AlGaN/GaN using line scans of images obtained at low magnification. Here we report on 2D-composition mapping in InGaN. This material is sensitive to electron beam irradiation as it leads to clustering of indium after a few minutes in the TEM using the parallel beam illumination mode [4, 5]. In contrast, we show here that InGaN is much more stable during STEM investigation under usual imaging conditions.
2. Experimental conditions
Our test sample was grown by metal organic vapour phase epitaxy (MOVPE) at temperatures in a range of 700 °C to 1100 °C. The process parameters were carefully chosen in order to minimize the indium fluctuations. The sample contains InGaN layers with a maximum In concentration of approximately 25%. STEM investigations were performed with a TITAN 80/300 TEM equipped with a Fischione Model 300 HAADF detector. The imaging parameters were as follows: Acceleration voltage 300 kV, spherical aberration constant of the probe forming lens $C_S = 1.2$ mm, radius of the condenser aperture 9 mrad, camera length 196 mm, HAADF detector angular range 33 to 200 mrad, gun lens 6, spot size 9, extraction voltage 4.5 kV, emission current 47 $\mu$A and $9.5 \times 10^{-11}$A probe current. Brightness and contrast were adjusted as described in reference [3]. For these settings of the illumination, the contrast transfer of the amplifiers is sufficiently linear [3]. Using a detector scan, we normalize intensities of STEM images with respect to the incident electron beam as suggested in [2]. The images consist of $2048 \times 2048$ scan points taken with a dwell time of 19 $\mu$s, resulting in a scan time of 80 s per image.

3. Image simulation
We used the STEMsim program [7] for simulation of the data needed for comparison with experiment. The simulations were performed for biaxially strained InGaN layers with In concentrations $< 50\%$ and specimen thicknesses $< 200$ nm with the frozen lattice approximation taking into account the nonhomogeneous sensitivity of our HAADF detector. Static atomic displacements were computed with the LAMMPS program [8] using the Stillinger-Weber parametrization published in reference [9]. Each computed STEM image was averaged over the area of one image unit cell and the results were tabulated in dependence of In concentration and specimen thickness.

4. Evaluation procedure
Figure 1a shows a small part of a STEM image that will be used to demonstrate the evaluation steps. The intensity of this image is normalized with respect to the incident electron beam [2, 3]. Figure 1b shows the same part of the image after noise filtering using a Wiener filter. To estimate the noise level, a circular area is chosen in the Fourier transformed image in such a way that it only contains noise. The noise level used for the Wiener filtering was chosen as 0.3 times the maximum absolute value found in the circular area. This Wiener filter is applied to facilitate detection of atomic column positions. For each bright spot in the image we search for the pixel with highest intensity. The position of this pixel is used as estimated position of the atomic column. Then, a local mean intensity is computed for each atomic column. To this end, the area of the image is segmented into two dimensional Wigner-Seitz cells. The highlighted area in Fig. 1c shows one of these cells as an example. Using the original non-Wiener filtered image, intensities of all pixels corresponding to one Wigner-Seitz cell are averaged which defines the local mean intensity (Fig. 1d). Then, the specimen thickness is evaluated for each atomic column within the GaN (blue region in Fig. 1d) by a comparison of the local mean intensity with image simulation. A polynomial is fitted to specimen thicknesses obtained in the GaN regions from which interpolated thickness values are obtained for the InGaN region of the image. Figure 1e gives the thickness map obtained for the example image. Finally, a comparison of local mean intensity (Fig. 1d) and evaluated specimen thickness (Fig. 1e) with the simulated data is used to obtain the In concentration evaluated for each atomic column. The resulting In concentration map is given in Fig. 1f.

5. Results and Discussion
Figure 2a and b show images 1 and 32 of a series of 32 images taken from the same region. Although the electron beam had scanned the same specimen area for 42 minutes after exposure
of image 32, there is no significant change of the contrast pattern. Fig. 2c displays a map of the In concentrations evaluated from image 1. Comparing Fig. 2c with d, it becomes obvious that no clustering of In can be observed after taking 32 images from the same area. An overview image that contained the illuminated area revealed a slightly increased background intensity most probably caused by carbon contamination. This leads to a decrease of the intensity contrast between GaN and InGaN, and thus to the reduction of the measured maximum In concentration from approximately 18 % in Fig. 2c to 16 % in Fig. 2d.

The small electron beam irradiation damage of InGaN we observed in our high resolution STEM experiments is in contrast to high resolution TEM, where In clustering is encountered after a few minutes [5]. A possible reason are different electron doses used in these experiments. In our STEM experiment, we used a probe current of $9.5 \times 10^{-11}$ A. For 80 s, this leads to a total dose on the investigated $50 \times 50$ nm$^2$ large area of 7.6 nC, resulting in a dose of 0.003 nC/nm$^2$. In reference [5], the electron beam current was reduced substantially below the maximum attainable from the microscope and was 35 A cm$^{-2}$. This led to In clustering after 220 s, corresponding to a dose of 0.08 nC/nm$^2$, which is significantly larger than the dose used in our STEM experiments.
Figure 2. a,b) Images 1 and 32 of a series of 32 STEM images obtained from the same specimen area. c,d) Color coded maps of the In concentration evaluated from the STEM images shown in the upper row. The legend shows the In concentration in percent.

However, after exposure of 32 images, the dose was 0.1 nC/nm² in our experiment, and no In clustering was observed. This indicates that the amount of In clustering not only depends on the electron dose, but can also be influenced by other parameters such as the growth method and the In-concentration [6].

6. Conclusion
We showed a procedure to measure 2D indium concentration maps from high resolution STEM images. We did not observe In clustering after scanning the same specimen area for 42 minutes. These findings are an important step towards investigation of indium clusters in as-grown samples, whose existence is a controversial issue.

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