Measurement of composition profiles in III-nitrides by quantitative scanning transmission electron microscopy

Journal of Physics: Conference Series **209** (2010) 012009 doi:10.1088/1742-6596/209/1/012009

Measurement of composition profiles in III-nitrides by quantitative scanning transmission electron microscopy

A Rosenauer[1], K Gries[1], K Müller[1], M Schowalter[1], A Pretorius[1], A Avramescu[2], K Engl[2] and S Lutgen[2]

[1] University of Bremen, Institute for Solid State Physics, Bremen, Germany
[2] OSRAM Opto Semiconductors GmbH, Regensburg, Germany

E-mail: rosenauer@ifp.uni-bremen.de

Abstract. In this paper we demonstrate a quantitative method for composition evaluation based on comparison of normalized image intensity with simulations carried out with the frozen lattice approximation. The method is applied to evaluate composition profiles of $Al_xGa_{1-x}N$/GaN layers. We measure ratios of image intensities obtained in regions with unknown and with known Al-concentration x, respectively. We show that estimation of specimen thickness combined with evaluation of intensity ratios allows quantitative measurement of composition profiles. Delocalization effects at interfaces due to instrumental resolution and dynamic electron diffraction are simulated. These effects can well be described by convolution with a Lorentzian. Measured intensity profiles can be corrected for delocalization effects using statistical parameter estimation so that deconvolution is avoided.

1. Introduction

Optical and electronic properties of light emitting diodes and laser diodes are governed by morphology and composition of semiconductor heterostructures. Therefore, research as well as quality control in industrial production require analysis of elemental concentration at high spatial resolution. Scanning transmission electron microscopy (STEM) using a high-angle annular dark field (HAADF) detector allows chemically sensitive imaging. We suggest quantitative analysis of composition based on comparison of normalized [1] measured detector intensity with image simulation [2] carried out with the frozen lattice or frozen phonon approximations [3].

2. Experimental procedures

STEM experiments were performed on a FEI TITAN 80/300 TEM/STEM equipped with an HAADF detector (Fischione Model 3000). Detector intensities are normalized with respect to the intensity of the incident electron beam similar to the method suggested by Le Beau et al. [1] but using the internal signal amplifiers of the TEM. Using different spot sizes as well as different settings of brightness and contrast of the amplifiers we found that good linearity is ensured for spot sizes between 6 and 11 if the detector dark signal is larger than zero and the signal corresponding to the incident electron beam is kept below $5 \cdot 10^4$. These values were measured by scanning the incident electron beam over the HAADF detector. The test structure investigated was grown by OSRAM with MOVPE and TEM specimens were prepared by focused ion beam (FIB) thinning.

3. Simulation of detector intensities

As measurement of specimen thickness and composition is based on a comparison with theoretical calculations, we simulated area scans for specimen thicknesses up to 200 nm and Al concentrations between 0 and 1 with the STEMSIM program [4]. The acceleration voltage was 300 kV, the spherical aberration of the probe forming lens was 1.2 mm, the convergence angle of the probe was 8.8 mrad and the detector covered an angular range between 33 mrad and 200 mrad. The non-homogeneous sensitivity of the detector was measured and taken into account. The simulations were carried out in the frozen phonon approximation for the $[2\bar{1}\bar{1}0]$ electron beam direction using a supercell with 7×7 unit cells. We obtained a converged calculation if the k-space ranged up to 400 mrad, corresponding to 1512×1596 pixels. The values for the mean square displacements were 3.24×10^{-3} Å^2 for Al and 3.59×10^{-3} Å^2 for N in AlN, as well as 3.47×10^{-3} Å^2 for Ga and 3.92×10^{-3} Å^2 for N in GaN, obtained by density functional theory. Linear combinations of mean square displacements were used for the ternary semiconductors. Each area scan covered one crystal unit cell and consisted of 20×20 calculation points. The detector intensity was then averaged over the image and plotted vs. the specimen thickness. The results are shown in figure 1. The accuracy of the GaN curve was checked with a FIB-lamella by comparison of specimen thickness evaluated from STEM Z-contrast images with the thickness directly measured in the FIB. We obtained an accuracy of the measurement of specimen thickness in GaN of ±15 nm.

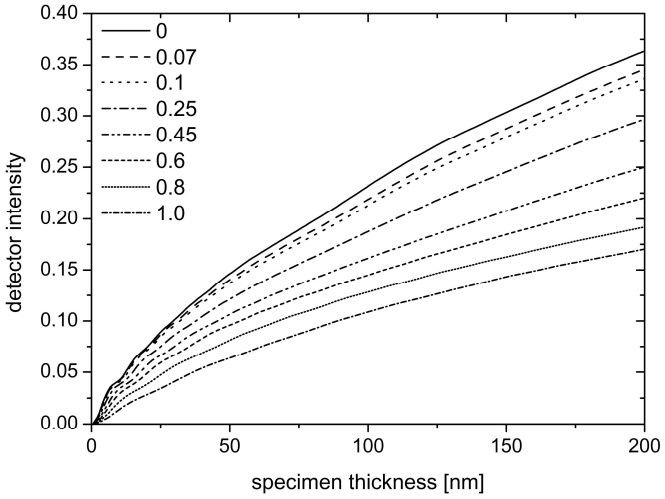

Figure 1. Simulated intensity averaged over one unit cell plotted vs. the specimen thickness. The curves have been computed with the frozen lattice approximation using 5 displacement configurations. The different curves correspond to the Al concentrations listed in the legend.

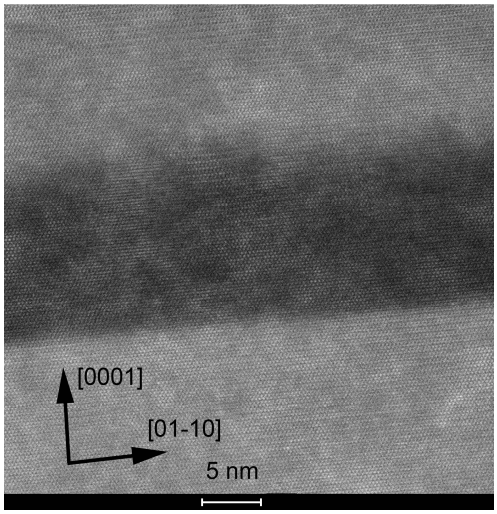

Figure 2. Experimental high-resolution STEM HAADF Z-contrast image of an $Al_{0.2}Ga_{0.4}N$ layer buried in GaN. The zone axis is $[2\bar{1}\bar{1}0]$ and the growth direction is [0001].

4. Experimental measurement and result

Figure 2 shows the experimental high-resolution STEM HAADF Z-contrast image whose evaluation is described in the following. First, we extracted the line scan shown in figure 3a, where we averaged along the $[01\bar{1}0]$ direction over a 20 nm wide region. The intensity oscillations mark the (0002) lattice planes. The intensity profile was Fourier transformed and the 0002 and 0001 Fourier components were deleted. The result was inverse Fourier transformed which led to the profile of the averaged image intensity shown as thick grey curve in figure 3a. Then we fitted a third-order polynomial to the GaN-regions. The resulting reference intensity is drawn as the dashed curve, corresponding to a hypothetical GaN crystal with the actual specimen thickness. Comparison with the solid curve in figure 1 yields the specimen thickness depicted in figure 3b. The intensity ratio shown in

figure 3c was calculated by dividing the normalized raw intensity by the normalized reference intensity. The Al-concentrations were computed for the local specimen thickness as given in figure 3b by comparing the profile of the experimental intensity ratio with simulated ratios obtained from figure 1. Positions of (0002) lattice planes were derived from the maxima positions of the intensity oscillations (black solid curve in figure 3a). Finally, the evaluated Al-concentration was averaged over half a unit cell for each (0002) lattice plane. The result is shown in figure 3d, where each box corresponds to one (0002) lattice plane. The composition profile indicates that the lower interface is sharper than the upper interface.

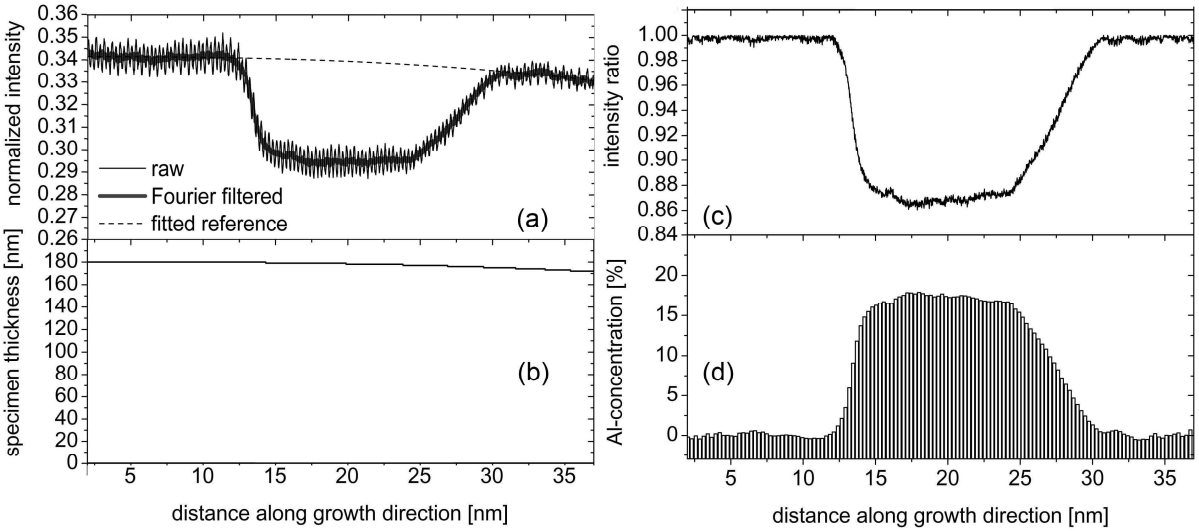

Figure 3. a) Normalized intensity obtained from averaging inside a 20 nm wide region in figure 2. The dashed curve is the reference intensity used for calculating intensity ratios as well as for evaluation of specimen thickness. b) Measured specimen thickness, c) intensity ratio and d) evaluated Al-concentration profile, where each bar corresponds to an (0002) lattice plane.

5. Effect of instrumental resolution and dynamic electron diffraction

5.1. Simulation

To check the influences of instrumental resolution and of a broadening of the electron beam by dynamic electron diffraction, we simulated an atomically sharp interface between GaN and $Al_{0.2}Ga_{0.8}N$ using the absorptive potential method. Figure 4 shows the obtained profile of normalized intensity. The maxima of the oscillation correspond to the positions of (0002) lattice planes. Then, the maxima positions and minima positions, respectively, were fitted by appropriate functions. The sum of maxima curve and minima curve divided by 2 gives the profile of the mean intensity indicated by the thick grey curve. This curve was used to calculate the delocalization function as demonstrated in figure 5. First, we defined an ideal intensity profile that contains a linear transition at the interface. This ideal profile was then convoluted with a Lorentzian $f(x) = 1/\left(\pi\delta^2\left[1+\left(x/\delta\right)^2\right]\right)$, where δ describes its width. The parameter δ was optimized so that the ideal profile convoluted with $f(x)$ fitted the simulated intensity profile.

5.2. Correction

The measured composition profile was corrected for delocalization effects without using deconvolution. For that, a model composition profile $m(x)$ was optimized so that $m(x) \otimes f(x) \otimes g(x)$ shows minimum deviation from the measured composition profile depicted in figure 3d. The function $g(x)$ is a Gaussian that describes the source size dependent broadening. Its half-width at half maximum value of 0.07 nm has been measured as described in [5]. Figure 6 shows

the corrected composition profile in comparison with the uncorrected profile for a small region containing the lower interface visible in figure 2. As the width δ is only 0.13 nm at 180 nm specimen thickness, the correction leads to small changes of the concentration profile only.

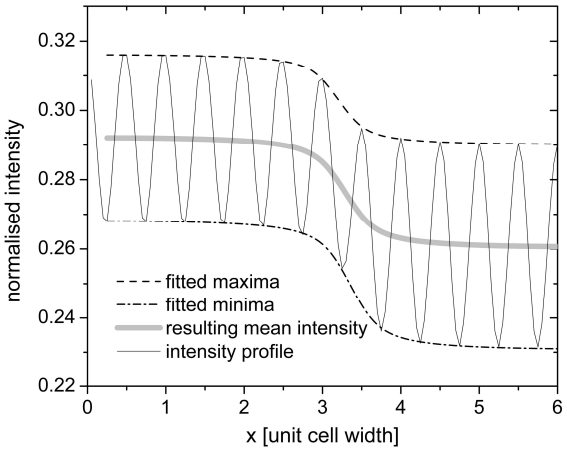

Figure 4. Profile of normalized intensity simulated for an atomically sharp interface between GaN (left) and $Al_{0.2}Ga_{0.8}N$ (right). The mean intensity profile was obtained as the mean of the fitted maxima and minima curves. The specimen thickness was 180 nm.

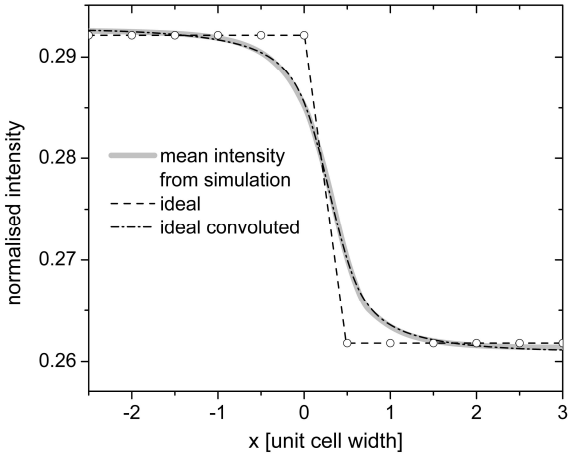

Figure 5. The dashed line shows the ideal intensity profile. The thick grey curve is the profile of the mean intensity shown in figure 4. The dash-dotted curve was obtained by convoluting the ideal profile with a fitted Lorentzian delocalization function.

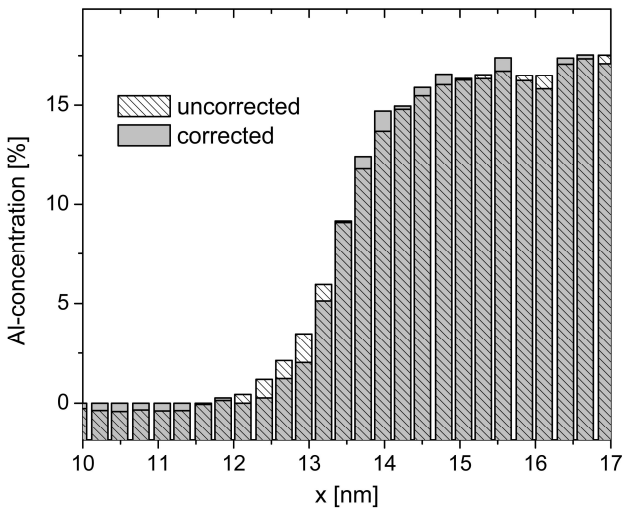

Figure 6. Corrected and uncorrected Al-concentration profiles plotted vs. the distance along the growth direction. Each bar corresponds to one (0002) lattice plane. The hatched bars show the original profile as depicted in figure 3d. The grey bars correspond to the model concentration profile $m(x)$, whose convolution with $g(x)$ and $f(x)$ yields the uncorrected Al-concentration profile. The function $g(x)$ describes the source-size dependent broadening and $f(x)$ is the delocalization due to instrumental resolution and dynamic diffraction of the electron beam in the specimen.

Acknowledgements
This work was supported by the Deutsche Forschungsgesellschaft under project II-2 of the research group FOR 506 and by project RO2057/4-1. A.R. thanks S. Stemmer for valuable discussion.

References
[1] LeBeau JM, Stemmer S 2008 Ultramicroscopy **108** 1653
[2] Grillo V, Carlino E, and Glas F 2008 Phys. Rev. B **77** 054103
[3] Rosenauer A, Schowalter M, Titantah JT, Lamoen D 2008 Ultramicroscopy **108** 1504
[4] Rosenauer A, Schowalter M 2007 Springer Proceedings in Physics **120** 169
[5] Rosenauer A, Gries K, Müller K, Schowalter M, Avramescu A, Engl K, and Lutgen S, submitted to Ultramicroscopy