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A (S)TEM and atom probe tomography study of InGaN

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Abstract. In this work we show how the indium concentration in high indium content $In_xGa_{1-x}N$ quantum wells, as they are commonly used in blue and green light emitting diodes, can be deduced from high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) images. This method bases on introducing normalized intensities which can be compared with multislice simulations to determine the specimen thickness or the indium concentration. The evaluated concentrations are compared with atom probe tomography measurements. It is also demonstrated how the quality of focused ion beam prepared TEM-lamellas can be improved by an additional etching with low energy ions.

1. Introduction

$In_xGa_{1-x}N$ is a well suited material for opto-electronic devices such as light emitting diodes and laser diodes, due to its variable bandgap which allows for light emission over the whole visible and part of the ultraviolet spectral range. Despite a high dislocation density leading to non-radiative recombinations, $In_xGa_{1-x}N$ based LEDs or laser diodes show a very good lighting efficiency. The reason for this is still under discussion, but small fluctuations of the indium concentration [1] or the layer thickness [2] are assumed to cause bandgap minima which work as traps for carriers and thus prevent them from moving to the dislocations. The precise determination of the indium content in such structures is still challenging. In this contribution we will introduce a method how the indium concentration can be directly derived from HAADF-STEM images.

2. Investigated specimen and optical properties

The investigated specimen was an InGaN/GaN test structure consisting of high indium content $In_xGa_{1-x}N$ quantum wells separated by GaN barriers. It was grown by metalorganic vapour phase epitaxy (MOVPE) on a low defect-density GaN substrate. Optical properties of the specimen were investigated via µ-photoluminescence (µ-PL) at a temperature of 4 K (figure 1). Beside typical contribution of the GaN barriers to the PL spectrum as the D^0X (3,469eV) and the LO-phonon replica the PL maximum of the spectrum is centred at 525 nm (2.36 eV). This bandgap energy corresponds to an In concentration of 29% for bulk material InGaN.

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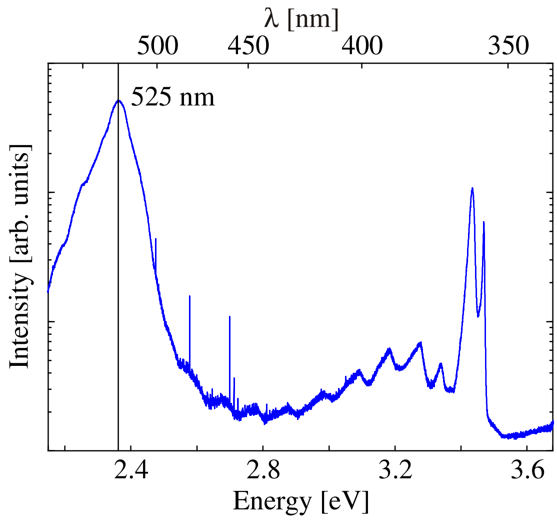

Figure 1. μ-PL spectrum with maximum at 525nm measured at 4K.

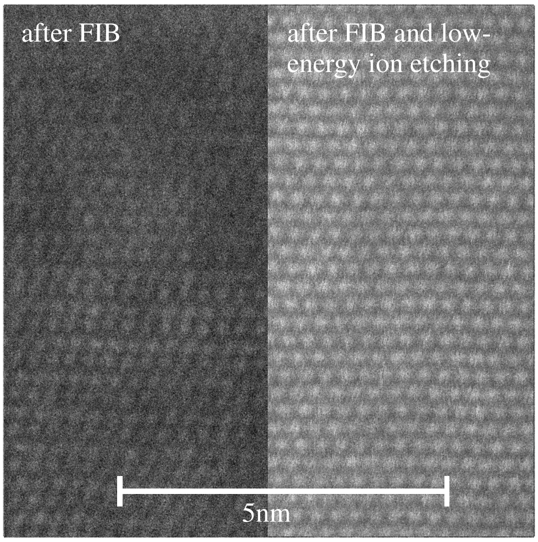

Figure 2. HAADF-STEM images before and after low-energy ion etching.

3. HAADF-STEM

3.1 Optimisation of preparation process

In recent years the FIB-liftout technique [3,4] has become one of the most common preparation methods for TEM-samples. Unfortunately, this procedure induces an amorphous surface layer, which leads to a stained image contrast and reduces resolution (figure 2 left side). Especially if specimens are investigated for concentration fluctuations or interface sharpness this is not desired.

We have minimised the amorphous surface layer by applying another etching step after FIB-preparation with low-energy ions ($E < 1$keV) using a Technoorg Linda GM IV5 system. The right side of figure 2 shows the best result that was found for an ion energy of 400eV leading to a reduction of the amorphous surface layer and thereby to a higher resolution. Furthermore, it is easier to identify concentration fluctuations.

3.2 Specimen thickness and concentration determination

To determine the indium concentration in the quantum wells HAADF-STEM images were taken with an FEI Titan 80-300 operated at 300kV. The integration angle of the detector was set to 33-200mrad. Before all measurements the detector was scanned in STEM imaging mode without any specimen inserted into the beam. This is done to adjust the detector in such a way that it shows a linear amplification characteristic and to measure the intensity of the scanning electron probe.

The probe intensity is used to normalize the measured image intensities following LeBeau and Stemmer [5], which allows quantitative analysis. These normalized intensities are afterwards compared with reference values simulated with multislice calculations in the frozen lattice approach using the STEMsim software [6]. Static atomic displacements were taken into account and the simulations were carried out in dependence on specimen thickness and indium concentration (figure 3). The specimen thickness is deduced by comparing the normalized intensities in regions of known concentration (GaN regions in this case) with the reference values for the according concentration (0% indium in this case) [7]. The specimen thickness is then interpolated over the regions of unknown concentration (figure 4).

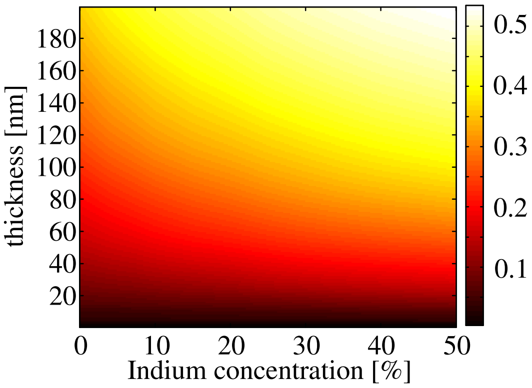

Figure 3. Reference values obtained via multislice calculations.

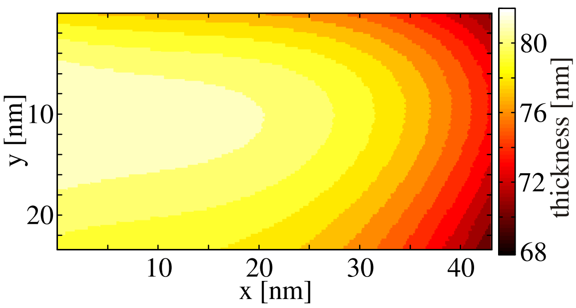

Figure 4. Specimen thickness derived from GaN regions and interpolated over the image.

To evaluate the indium concentration, the normalized intensities in quantum wells are compared with the reference values, this time taking the interpolated thickness of figure 4 into account. This leads to the calculated In concentration map shown in figure 5. The indium content of the two shown layers increases up to values around 23%. Averaged along the monolayers in growth direction ((0002)-planes) an average concentration of (17.7±1.7)% for the first (QW1) and (18.9±1.8)% for the second quantum well (QW2) can be found for this image, but these values fluctuate over the specimen. We observe regions where the quantum wells are interrupted and regions where they possess a lower concentration.

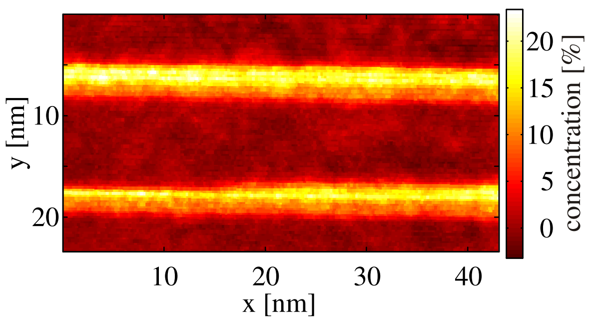

Figure 5. Indium concentration map (HAADF-STEM).

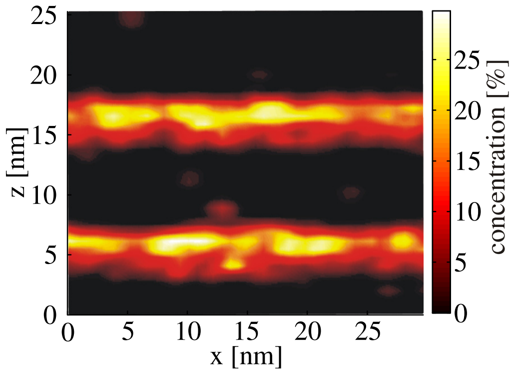

Figure 6. Indium concentration map (Atom probe tomography).

4. Atom probe tomography (APT)

APT measurements were carried out on a laser assisted Imago LEAP 3000 system and the specimen was reconstructed with the IVAS software. The reconstruction was analysed for the average indium concentration and for small-range indium fluctuations. For the average indium concentration all atoms within bins of 0.1nm lying in planes perpendicular to the growth direction were taken into account. A maximum indium concentration of around 25% was measured, which is significantly higher than the values obtained from HAADF-STEM. Most probable reasons for this deviation are long range fluctuations of the In concentration that lead to a decrease of the average composition in projection, and the broadening of the electron beam within the specimen.

Figure 6 shows a concentration map of one reconstruction averaged over a thickness of 1nm. There seem to be indium clusters with a diameter of around 5nm present, especially in the bottom layer. In these clusters the indium concentration increases to nearly 30%. This is in good agreement with the concentration calculated from the PL spectrum. However, it is not quite sure if these concentration fluctuations are not only due to statistical fluctuations in the ternary compound.

5. Conclusion

In this work an InGaN based multi quantum well test structure was investigated via HAADF-STEM and APT. It was shown how the indium concentration and specimen thickness can be directly deduced from HAADF-STEM images by introducing normalized intensities. The concentrations measured via HAADF-STEM are significantly lower than the results obtained via APT and calculated from the PL spectrum. This may be due to long-ranging fluctuations in the indium concentrations of the quantum wells and layer interruptions which were observed but not shown here. Especially if these fluctuations are present in electron beam direction, they could be responsible for the discrepancy.

The atom probe reconstructions were also investigated for fluctuations on smaller length scales. Indium clusters with a diameter of around 5nm seem to be present, but it is still an open question if they are only of statistical origin.

Acknowledgements

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References

[1] Chichibu S, Sota T, Wada K and Nakamura S 1998 *J. Vac. Sci. Technol.* B **16** 2204-14

[2] Graham D M, Soltani-Vala A, Dawson P, Godfrey M J, Smeeton T M, Barnard J S, Kappers M J, Humphreys C J and Thrush E J 2005 *J. Appl. Phys.* **97** 103508

[3] Overwijk F M H, van den Heuvel F C and Bulle-Lieuwma C W T 1993 *J. Vac. Sci. Technol.* B **11** 2021-24

[4] Giannuzzi L A and Stevie F A 1999 *Micron* **30** 197-204

[5] LeBeau J M and Stemmer S 2008 *Ultramicroscopy* **108** 1653-58

[6] Rosenauer A and Schowalter M 2007 STEMsim - a New Software Tool for Simulation of STEM HAADF Z-Contrast Imaging *Microscopy of Semiconducting Materials 2007* ed A G Cullis and P A Midgley (Springer Netherlands) pp 170-172

[7] Rosenauer A, Gries K, Müller K, Pretorius A, Schowalter M, Avramescu A, Engl K and Lutgen S 2009 *Ultramicroscopy* **109** 1171-82