Measuring composition in InGaN from HAADF-STEM images and studying the temperature dependence of Z-contrast

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Abstract. In this contribution, the indium concentration profile of an InₓGa₁₋ₓN/GaN five-fold multi quantum well structure is measured from high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM) images. The results are compared with an atom probe tomography study. Indium concentrations in the range of 26 at.% to 33 at.% are measured in the centre of the quantum wells. An additional indium layer of 14 at.% has been found on top of the quantum wells. In the second part, the temperature dependence of measured intensities in GaN is investigated. Here, multislice calculations in the frozen lattice approximation are carried out in dependence of specimen thickness and compared to experimental data. An increase of intensity with specimen temperature is found.

1. Introduction

Semiconducting materials are used in a variety of technological applications due to their electronic properties. For example, InₓGa₁₋ₓN with its bandgap ranging from 0.7eV [1] to 3.5eV [2] is particularly suitable for optoelectronic devices such as light emitting diodes or laser diodes that operate in the visible spectrum of the light. As a typical LED contains several layers of different semiconducting materials and layer thicknesses of only a few nanometres, adequate techniques for composition analysis at sub-nanometre scale are required.

Nowadays, in scanning transmissions electron microscopes (STEM) a high spatial resolution well below one nanometre can be achieved and in combination with a high-angle annular dark field (HAADF) detector the image intensity also strongly depends on the atomic number (Z) of the scattering atoms. Thus, HAADF-STEM is an appropriate technique for composition analysis.

Rosenauer et al. [3] demonstrated on an AlGaN/GaN heterostructure that specimen thickness and material composition can be deduced by a comparison of measured intensities with simulated reference data. The comparison is based on a normalisation of image intensities with respect to the scanning probe [4]. Grillo et al. [5] pointed out that static atomic displacements (SAD) due to different covalent radii of different atomic species have to be considered in multislice calculations for an accurate simulation of HAADF intensities.

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2. Experimental Setup

The investigated specimen was a five-fold InGaN/GaN multi-quantum well (MQW) structure grown by metal-organic vapour phase epitaxy (MOVPE) in a horizontal reactor (Aictron AIX 200RF). The InGaN quantum wells were separated by 20 nm GaN barriers (more details on specimen growth can be found in the publications of Rossow et al. [6] and Hoffmann et al. [7]). For the STEM analysis, a thin lamella was prepared by focused ion beam lift out. Due to the preparation process, the specimen surface is usually covered with an amorphous layer of a few nanometres. Additional etching with argon ions of low energy (400 eV) has been applied to remove this layer [8]. STEM investigations were performed with an FEI TITAN 80/300 equipped with an HAADF-detector (Fischione model 3000). A spot size of 6 and a gun lens setting of 9 was used. The integration angle of the HAADF-detector ranged from 36 to 220 mrad.

Atom probe tomography (APT) was conducted on an Imago LEAP™ 3000X HR system. Here, the analysed specimen was also prepared by focused ion beam lift-out. To gain a III:V ratio close to 50:50 the specimen was cooled to 40K and the energy of the triggering laser pulses was reduced to 0.05 nJ.

3. Composition analysis

For the quantification, reference intensities were simulated for In$_x$Ga$_{1-x}$N with indium concentrations ranging from $x = 0$ to $x = 0.55$ and specimen thicknesses up to 150 nm. These multislice calculations in the frozen lattice approach were performed using the STEMsim program [9]. The simulations included SADs [5] and the non-uniform sensitivity of the HAADF-detector was also taken into account. A parameterisation of the reference data and a more detailed description of the simulation can be found in [10].

Figure 1 displays an HAADF-STEM image of the five quantum wells. The indium concentration was extracted by normalizing the measured intensities with respect to the intensity of the scanning electron probe [4]. Afterwards, the intensities of the GaN barriers were interpolated over the quantum wells and used to determine the specimen thickness by comparing them with the simulated data for GaN. Then, the intensities measured within the quantum wells were compared with the reference data taking the interpolated thickness into account. This yielded the concentration profile shown in figure 2. Concentrations between 30 and 33 at.% indium were measured within the quantum wells and 14 at.%
Indium were detected within an additional layer on top of the wells that formed during specimen growth. For comparison, the specimen was also analysed by APT (see lower part of figure 1). The deduced concentration profile is also shown in figure 2. Here, concentrations in the range of 26 to 31 at.% were measured within the quantum wells and of 13 at.% in the additional layers.

Figure 3. Simulated HAADF intensity of GaN in dependence of temperature.

Figure 4. Intensity profiles for different temperatures. The MQW-region corresponds to figure 1.

4. Temperature dependence of HAADF-intensity

A main contribution of the intensity scattered into high angles originates from thermal diffuse scattering due to thermal vibrations of the specimen atoms. In order to investigate the influence of the specimen temperature on the measured intensities frozen lattice simulations were performed for GaN for varying specimen temperature. The specimen temperature was included into the simulations by temperature dependent Debye-Waller factors [11].

In figure 3 the simulation results are shown for specimen temperatures of 300 K, 450 K and 600 K in dependence of the specimen thickness. As can be seen, the intensity increases with increasing specimen temperature due to the higher thermal displacements of the specimen atoms.

This has also been observed experimentally. The specimen was heated in a Gatan double tilt heating holder (Model 652) and HAADF-STEM images were acquired at exactly the same position. Linescans were extracted that are shown in figure 4. For the study of the temperature dependence of the GaN intensities, the GaN region below the MQW-region has been used. The general tendency of all three profiles to increase from the left to right is due to increasing specimen thickness. Nevertheless, it can be also seen that the measured intensities increase with temperature. As expected from the simulations, the difference between the three profiles is increasing with increasing specimen thickness and the difference between the 300 K and 450 K profiles is larger than the corresponding difference between the 450 K and 600 K curves.

5. Conclusions

We have demonstrated the composition measurement by quantitative HAADF-STEM on an InGaN/GaN heterostructure. An average concentration (QW1-QW4) of (31.9±1.0) at.% indium has been measured that is in well agreement with concentrations measured by an atom probe tomography study (30.5±0.9) at.%.
In the second part, the temperature dependence of HAADF-STEM has been studied for GaN. An increase of the measured intensities has been found, which was also predicted by multislice calculations. The increase of the intensities is caused by an increase of thermal diffuse scattering that rises the amount of electrons scattered into higher angles.

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