Quantitative measurement of composition fluctuations in InGaN quantum wells

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Measuring the local lattice parameter from high resolution transmission electron microscopy (TEM) phase contrast images is a common method to determine the local composition in a semiconductor alloy. Under the assumption that (i) the information in the image is local, i.e. the contrast pattern corresponds to the projected atomic potential and (ii) based on the knowledge of the elastic parameters and a strain model (uniaxial or biaxial depending on the TEM sample thickness) one may conclude from the local lattice parameter on the composition. The precision in measurement of lattice parameters and thus the compositional sensitivity of the measurements depends on systematic and statistical errors. While systematical errors such as lens aberrations can be essentially reduced in an aberration corrected microscope, statistical errors pose a serious limitation on the sensitivity. For example to measure an uniaxial strained InGaN alloy with a compositional sensitivity of 1% requires a precision of 0.75 pm. Two sources of experimental error are of particular importance (i) the finite signal-to-noise ratio (SNR) of the detection system and (ii) the influence of amorphous surface layers. As long as the noise has only minor frequency dependency especially at frequencies of the crystal lattice, the lattice parameter measurement precision will not be degenerated. From image simulations of GaN with similar conditions as in the experiment, added white noise (no frequency dependency) with a SNR of 20 leads to a standard deviation (STD) in the lattice parameter measurement of 0.68 pm which is significantly smaller than the typical STD of around 5 pm found in experiments [1]. A more serious limitation comes from the hardly avoidable “noise” due to amorphous surface layers. These layers are caused either by ion milling or adsorbents and have a frequency spectrum, that is governed by the radial distribution function in the amorphous layer. When using the the contrast of an amorphous layer at the edge of the TEM sample from an experimental as a model for image simulation we obtain a STD of 4.7 pm for the measured lattice parameter, which is close to the experimental STD value and suggests that these amorphous layers are indeed an important error source for the lattice parameter determination. One way to reduce the error is averaging over a bigger area at the cost of local resolution as recently shown in [2].

In the present paper we will propose an experimentally simple and robust method that considerably improves the precision for the lattice parameter determination from HRTEM images despite the presence of amorphous surface layers and without any loss of spatial resolution. As amorphous layer are not stable under the electron beam as can be seen by the changing contrast of these layers we use image series recorded under the same conditions e.g. without changing the defocus as in focus series used for exit wave reconstruction. Each image of the series is evaluated in the same way as in single image analysis. We use the peak finding method with sub-pixel position estimation using an iterative centre-of-gravity approach and measure the peak distances to find the local lattice parameters. Then the measured distances are averaged throughout the series to give the final result. Due to the sample drift during the acquisition time of the series one has to identify the same peak throughout the series which can be easily achieved in the chosen approach without the need of sub-pixel alignment of the images.

Using the image series approach we investigated an $In_xGa_{x-1}N$ multi quantum well (QW) structure consisting of 4 quantum wells which are grown at 750°C onto a 4 µm GaN layer on a sapphire substrate by metal organic chemical vapour phase deposition. The $In_xGa_{x-1}N$ quantum

wells are step graded, i.e. each quantum well consists of two InGaN layers, a thin $In_{0.09}Ga_{0.94}N$ layer, followed by an $In_{0.16}Ga_{0.84}N$ layer with a total thickness of about 3 nm. The quantum wells show a room temperature photo luminescence (PL) peak position at 450 nm consistent with the intended composition of 16% Indium. Dislocation densities were in the range of 5 x $10^{8}cm^{-2}$, as determined from plan view TEM images. Cross-sectional samples were prepared using mechanical polishing (diamond foils) down to a grain size of 0.5 µm. Final preparation was done by ion milling with acceleration voltages from 4 keV to 0.2 keV. With this preparation technique, we typically achieve a wedge shaped sample with a thickness from about 4-5 nm close to the edge. The TEM images were required with an aberration corrected FEI Titan 80-300 microscope operated at 300 kV. For imaging, we tuned the spherical aberration to a slightly negative value (-12 µm) and record the images with a slight overfocus (+6 nm). As was first shown by Lentzen et al. [3], this enhances the phase contrast and reduces contrast delocalization to values below the information limit of the microscope, which is at 0.08 nm (measured by the Young fringe test). Under these conditions, atomic columns appear as bright dots on dark background and resemble the projected atomic potential. The samples are oriented in the [1100] direction. In this direction the contrast pattern shows weak dependence from defocus and thickness changes below the first extinction length.

In Fig. 1 the measured local c-lattice parameters from a single image is shown next to the averaged result of the series. As can be seen the variation in the result of the averaged series is reduced compared to the single image evaluation and the staggered QW structure is now clearly resolved. In the GaN region the STD of the measurement drops from 5.3 pm to 1.5 pm without loosing spacial resolution. The mean In content in the upper and lower part of the QW is found to be about 16 % and 9 % respectively assuming an uniaxial strain state for the TEM sample with a thickness of around 8 nm. The improved precision in the measurement allows a statistical analysis of the local measured values. With a STD of 2.9 pm in the upper part of the QW for the c-lattice parameter the result shows no evident discrepancy to a random alloy, e.g. no hint for In clustering in the QW.

References

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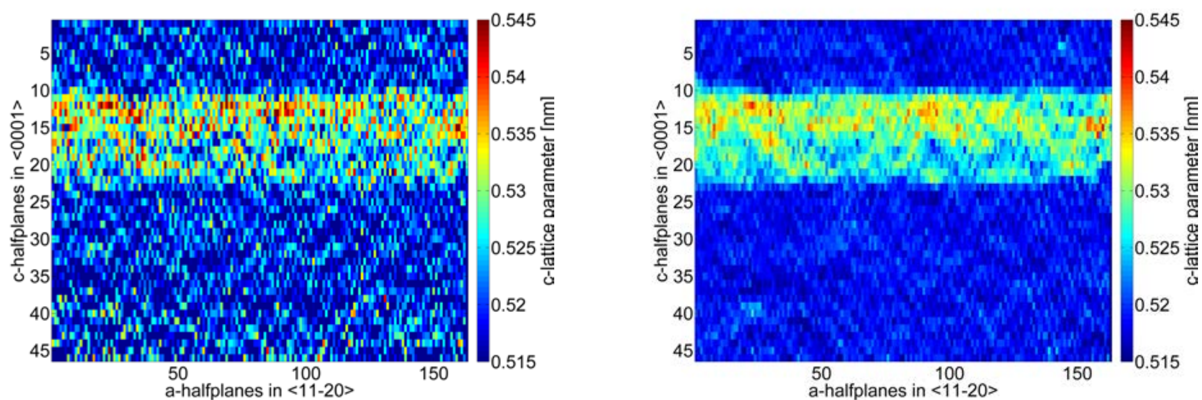

Figure 1. *Single image evaluation of the local c-lattice parameter (left) compared to the evaluation of the series of 30 images (right) of a staggered InGaN quantum well.*