Quantitative High-Resolution TEM/STEM and Diffraction

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Determination of In and N concentration in (InGa)(NAs) quantum wells using HAADF STEM and investigation of annealing effects

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The zincblende crystal $In_xGa_{1-x}N_yAs_{1-y}$ is of technological interest as both, In and N lower the semiconductor's band gap, allowing to emit or to absorb in the infra-red, so that InGaNAs can either be used for laser diodes in telecommunication at wavelengths of 1.3 and 1.55 □m - here glas fibres show optimum transmission - or as part of multi-junction solar cells. It is known that thermal annealing of InGaNAs in order to achieve an increase of the emission intensity typically also causes an unwanted blue shift of the emitted wavelength. In this contribution we present a method to simultaneously determine both, the In and the N concentration, from high-angle annular dark field (HAADF) micrographs recorded in high-resolution (HR) scanning transmission electron microscopy (STEM). For this, two sources of information are extracted on atomic scale: on the one hand the distances of atomic columns are measured which allows for a strain state analysis, on the other hand the characteristic HAADF intensity is evaluated by a comparison to simulated intensities. HAADF-intensity evaluation was already shown for (e.g.) InGaN [1] and GaNAs [2]

For the simulations a frozen-lattice multislice approach is used in the STEMsim software [3]. Besides thermal diffuse scattering (TDS), Huang-scattering at static atomic displacements (SADs) is taken into account [4]. SADs are distortions of the atomic lattice due different covalent radii of In and Ga as well as As and N. We compute these static displacements by relaxing the supercells with the LAMMPS [5] package using Keating's valence force field model [6]. Figure 1. shows the ratio of the simulated HAADF intensity of InGaNAs and GaAs as a function of specimen thickness for different compositions (see Figure 1.). The high concentration dependent contrast allows for an accurate determination of the chemical composition. An MOVPE grown InGaNAs/GaAs quantum well sample (*as-grown*) is characterized in [100] zone axis by the suggested method. Figure 2.(a) shows the concentration maps for In and for N, Figure 2(b) the concentration profile from averaging along [010]-direction (red: In, green: N). The mean concentration is determined to be 32 % of In and 2 % of N. These values are in good agreement with the results from high-resolution X-ray diffraction measurements (HRXRD). Atom-probe tomography (APT) was applied to the sample. The corresponding In profile is also shown in Figure 2(b). Both, profile shape and mean concentration, is in good agreement with the results from the HAADF image.

The evaluations (HAADF-STEM analysis, HRXRD and APT) were also applied to a sample of the same growth series which was annealed under As-stabilized conditions. We found that the composition profiles did not change, but photoluminescence showed a blue shift of approximately 60 meV. This blue shift can be explained by a local redistribution of N atoms from Ga-rich to In-rich sites [7, 8].

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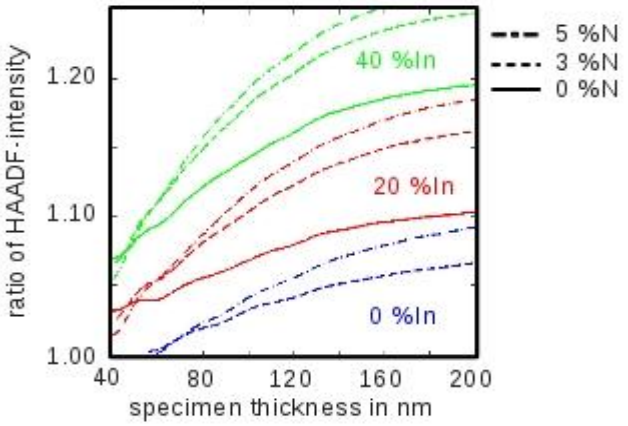

Figure 1. Ratio of the simulated HAADF intensity for InGaNAs and GaAs (contrast) as a function of specimen thickness. Each curve displays a different indium concentration (color) and nitrogen concentration (line style).

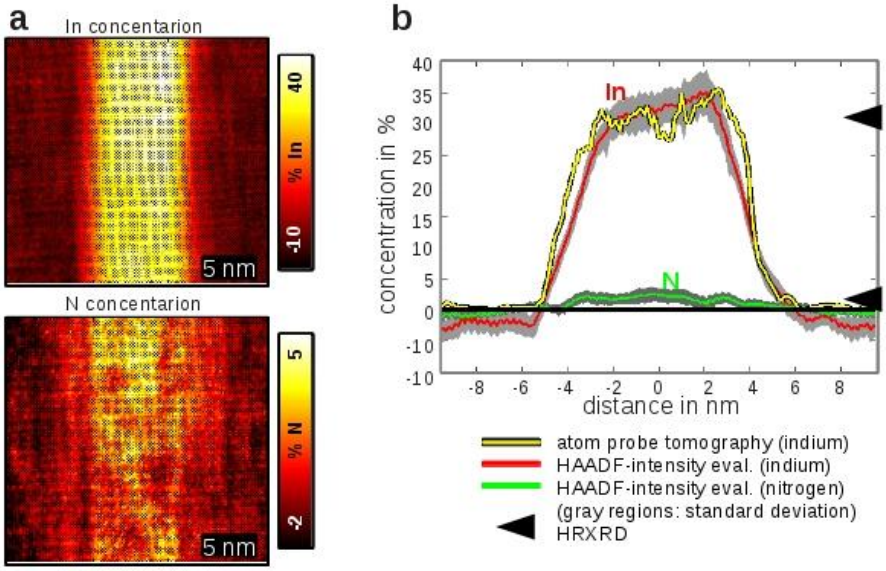

Figure 2. (a) Concentration maps from HAADF-intensity evaluation for indium and nitrogen. (b) Profiles from averaging the maps in (a) and from atom probe tomography. Concentrations derived from HRXRD are marked by arrows.