InGa\textsubscript{1-x}N\textsubscript{x},As\textsubscript{1-y} is of technological interest for laser diodes in telecommunication and solar cells as both, In and N, lower the semiconductors band gap to emit or absorb in the infra-red spectral range. It was shown for ternary materials that an unknown chemical concentration (e.g. of In in InGaN \cite{1}) can be determined by high-angle annular dark field (HAADF) scanning transmission electron microscopy (STEM). For this purpose experimental HAADF intensities are compared with simulated ones. The experimental intensities are normalized to the total beam intensity which allows for determining the thickness in regions with known chemical composition. In this contribution this method is extended to evaluate the quaternary system InGa\textsubscript{1-x}N\textsubscript{x},As\textsubscript{1-y}. As a specific HAADF intensity cannot be allocated to a pair of concentrations (x,y) in a unique way, further information is needed. To this end, the local strain state is additionally determined from the high-resolution HAADF-STEM image.

The HAADF intensities were simulated with a frozen-lattice multislice approach implemented in the STEMsim software \cite{2}, considering thermal diffuse scattering (TDS). It was shown that for (In)GaNAs, besides TDS, Huang-scattering at static-atomic displacements (SADs) has to be taken into account \cite{3}. SADs are distortions of the atomic lattice due to different covalent radii of In and Ga as well as As and N. The SADs were computed by relaxing the supercells using Keating's valence force field parametrization \cite{4} in the LAMMPS code \cite{5}. Fig. 1. shows the ratio of the simulated HAADF intensity of InGaNAs and GaAs versus specimen thickness for different In and N concentrations. For thicknesses above approx. 50 nm the intensity ratio increases not only with In but also with N concentration, although N has a smaller atomic number than As. This effect reveals the strong influence of additional scattering at SADs. An MOVPE grown InGaNAs/GaAs quantum-well sample is characterized by the outlined method. The mean concentrations of 32 \% In and 2 \% N (see concentration profiles in Fig. 2) are in good agreement with the results from XRD (marked by arrows). In addition, atom-probe tomography was applied to this sample, and the corresponding In profile is also shown in Fig. 2. Both, profile shape and mean concentration are in good agreement with the HAADF-STEM results.

\cite{1} Rosenauer et al., Ultramicroscopy 111 (2011) 1316.
\cite{4} P. N. Keating, Phys. Rev. 145 (1966), 637.

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Fig. 1: Ratio of the simulated HAADF intensity for InGaNAs and GaAs (material contrast) as a function of specimen thickness for different indium concentrations (color) and nitrogen concentrations (line style). The HAADF intensity increases for specimen thicknesses above 50 nm with In and N concentration due to Z-contrast and scattering at SADs.

Fig. 2: Determination of the chemical composition of an InGaNAs layer embedded in GaAs. Concentration profiles from averaging concentration maps (HAADF analysis: indium and nitrogen) and from atom probe tomography (only indium). Concentrations derived from HRXRD are marked by arrows.