Dual compositional mapping in InGaNAs using a single TEM lattice fringe image

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The analysis of InGaNAs solar cell and infrared laser structures has raised the interest of several physical disciplines, especially semiconductor optics, -epitaxy, solid state theory and transmission electron microscopy (TEM). One remaining challenge is to clarify the correlation between structural properties, e.g. the In- and N composition at atomic scale, and post-growth thermal annealing conditions with respect to the optical and spectral performance of InGaNAs nanostructures.

Here, we present a new TEM technique to measure both local In and N content at atomic scale and demonstrate the application to nanostructures with different composition ranges. Both strain and chemically sensitive 020 amplitude are measured from 220 and 020 fringes in one TEM three-beam lattice fringe image and compared with simulated reference data. In order to eliminate nonlinear contributions to the respective diffractogram reflections, an L-shaped objective aperture was fabricated by focused ion beam milling and inserted in a Titan 80/300 image corrected TEM. In this way, our method not only overcomes the problem of correlating different images containing strain and chemically sensitive contrast separately [1], but also extends the method proposed in [2] to concentrations with vanishing 020 fringe amplitude. Composition maps for two quantum wells are shown in FIG. 1. Imaging conditions to minimise the impact of unknown specimen thickness were derived from Bloch wave simulations, yielding an optimum Laue Circle center of (4.2 0 0).

Bonding and static atomic displacements (SAD) play a central role in simulations of TEM images in dilute nitrides. The former effect can be treated by density functional theory (DFT) as only small cells must be considered, the latter is of statistical nature and requires strain relaxation in large cells, e.g., by valence force field (VFF) methods. We present a detailed study of the reliability of atomistic scattering models, such as isolated [3] and modified [4] atomic scattering amplitudes (ASA), when SAD are involved. As shown in FIG. 2, VFF minimises atomic forces drastically. Furthermore, structure factors derived from modified ASA combined with VFF are in best agreement with full DFT results, offering an efficient way to include bonding and SAD in structure factors for large cells. Since SAD give rise to diffuse losses to the background in a diffraction pattern, we account for this issue by defining additional absorptive form factors analogous to [3] and verify this approach by comparison of Bloch wave and multislice results for the diffracted beams, as shown in FIG. 3.

References

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[5] This work was supported by the DFG under contracts RO 2057/4 and SCHO 1196/3.

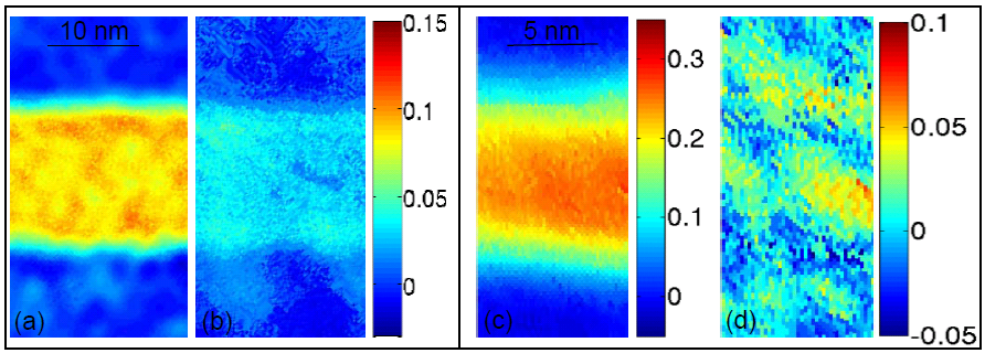

FIG. 1. Distribution of (a) In and (b) N in a lattice-matched solar cell structure and (c) In and (d) N in a laser structure. Each pair of maps was extracted simultaneously from one TEM lattice fringe image formed by 000, 220 and 020. A cosine was fitted to the 220 fringes, yielding fringe positions with subpixel accuracy, from which local strain is calculated. By Fourier filtering, the chemically sensitive local 020 fringe amplitude was obtained.

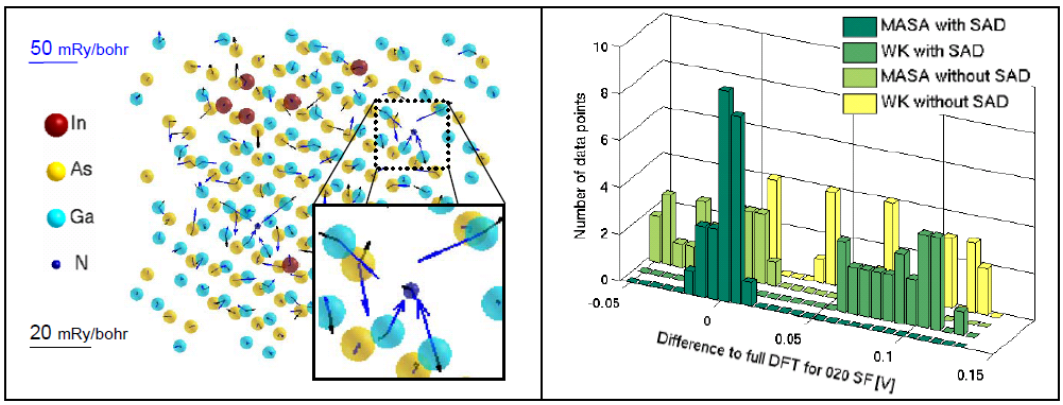

FIG. 2. *Left:* InGaNAs supercell with 216 atoms and atomic forces calculated by DFT. Blue and black vectors illustrate forces before and after VFF relaxation, respectively. *Right:* Structure factors were calculated by full DFT for 25 VFF relaxed supercells and compared with those obtained in isolated atom approximation (WK [3]) and using modified ASA (MASA [4]) with (dark green bars) and without (light green and yellow bars) SAD. The bars depict histograms of the differences to the full DFT result for the 020 structure factor. As desired, the data set for MASA with SAD is close to zero, whereas a neglect of either bonding or SAD can lead to large deviations from DFT.

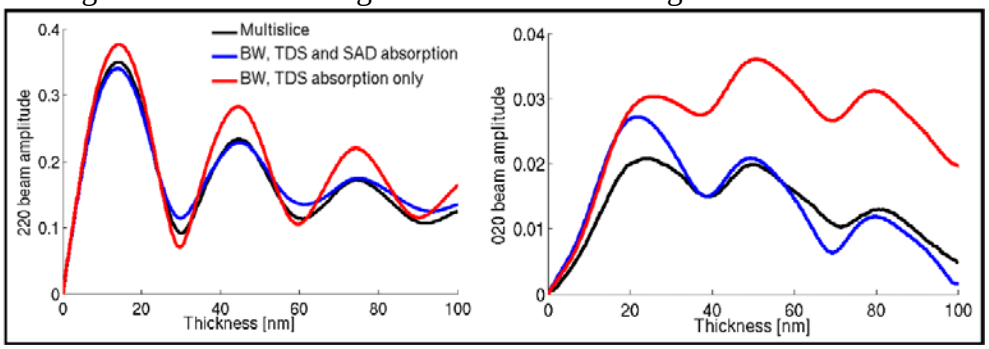

FIG. 3. Thickness dependence of the 220 (left) and 020 (right) beam amplitude for a VFF relaxed InGaNAs crystal containing 25% In and 5% N. The black curve depicts the multislice result. The Bloch wave (BW) simulations both rely on structure factors for the VFF relaxed cell according to [3]. For the blue curve, additional absorptive form factors were defined based on the mean SAD of each atomic species, yielding a significantly improved agreement with the multislice simulation.