Enhancing quantifiability of S/TEM-based composition mappings through correlative techniques

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Background incl. aims
In the realm of semiconductor physics and material engineering, the precise quantification of alloy concentrations is imperative as slight variations in the concentration can have far-reaching implications for the optical and electrical properties of semiconductor devices, e.g. InGaAsN near-infrared lasers [1]. As humanity continuously advances the frontiers of nanotechnology and device sizes shrink to just a few atomic layers, material's analysis must keep up and provide sufficient precision in the determination of alloy concentrations as well as the necessary spatial resolution to resolve these tiny devices. While S/TEM based approaches provide this necessary spatial resolution, each method used to determine elemental concentrations in STEM presents its own challenges, especially when it comes to quantifiability. To reliably determine concentrations, STEM-EDX for example, relies on proper reference samples and detailed knowledge of the systems x-ray signature [2]. Diffraction based techniques, such as dark-field imaging of a chemically sensitive beam, exhibits strong dependencies on the specimen's thickness and bending, as these influence dynamic diffraction conditions [3].

The study aims to improve the accuracy of compositions mappings by leveraging the individual challenges and advantages of EDX and diffraction-based techniques against each other.

Methods
We prepare a TEM-lamella, incorporating InGaAs layers (with known In concentration) near the [100] foil normal, employing classical cross-section preparation. This specific zone axis, characterized by 4 {002} reflections, exhibits chemical sensitivity as the structure factors of these {002} reflections hinge on the difference in atomic scattering factors within the unit cell of constituent materials. To exploit this chemical sensitivity optimally, we strategically tilt the specimen, predominantly exciting one of these reflections. This minimizes dynamic diffraction effects while concurrently enhancing the relative intensity of the respective reflection. Employing an ultrafast pixelated detector, we systematically scan the tilted specimen, capturing 2D diffraction patterns. In a complementary set of measurements, STEM-EDX signals from the layer are acquired for a comparative analysis. STEM-EDX measurements and 4D-STEM mappings are conducted on a JEOL ARM300F2 at 300kV with a 2.2 sr EDX-system. The integration of these measurements is achieved through 2D cross-correlation of the simultaneously acquired signals on the HAADF detector. Subsequently, virtual dark-field image of the (002) reflection is computed, facilitating a direct correlation with the structure factor and, consequently, revealing the corresponding Indium concentration within the layer. Results are compared to Bloch waves simulations to account for dynamic diffraction effects for the respective lamella thickness. Further investigations of the Indium segregation are conducted in high-resolution conditions analyzing the geometric phase.

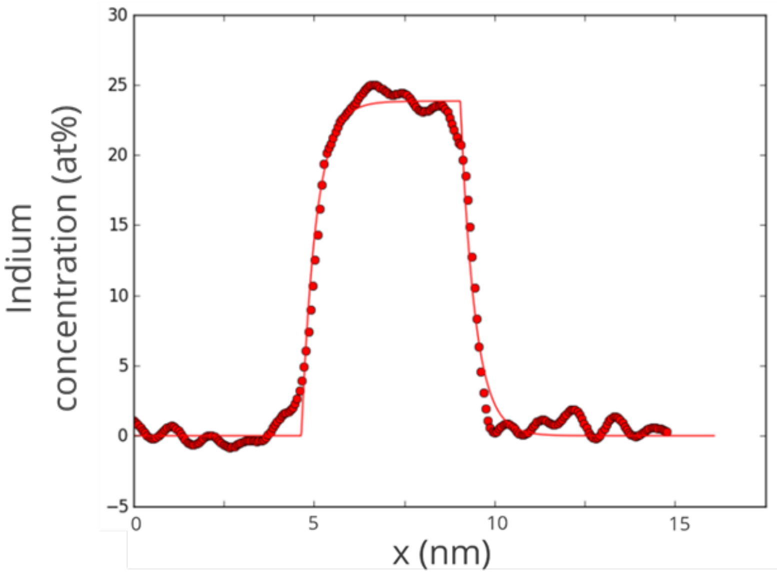
Results

From a dark-field image of the (002) reflection, a segregation profile for the Indium concentration within a 5 nm layer of InGaAs, nominally possessing a 20% indium concentration, can be extracted. It's noteworthy that, for the specified indium concentration and under strong-beam conditions, no significant disparity in indium levels, attributable to dynamic diffraction, is noticeable when comparing the (020) and (002) reflections. In Fig. 1 a segregation profile of the Indium concentration with a Muraki-Fit [4] is shown. Furthermore, for small Indium concentrations, the dependency of the concentration determined by this dark-field imaging on the specimen thickness seems to be negligible. This assertion finds support in Blochwave simulations, analyzing dynamic diffraction. For larger Indium concentrations, strain effects play an increasingly important role in the analysis of concentrations through dark-field imaging. Here, we explore the possibilities of determining concentration profiles from strain analysis, taking into consideration strain relaxation at the lamella surfaces. The obtained results are compared to STEM-EDX mappings of the same quantum well layer. Our comprehensive discussion covvers the results and their implications for the interplay with specimen thickness, Indium segregation, and how these factors collectively impact spatial resolution and quantifiability in STEM-EDX. In further investigations, we replicate the study using a set of TEM specimens prepared through a FIB TEM-lamella preparation. This replication aims to scrutinize the influence of FIB Ga-ions on the composition determination in Ga-based heterostructures, the changes to the In-distribution and amorphized layers.

Conclusion

Among the multitude of interaction possibilities of the high-energy electron beam, traditional approaches often focus on a single interaction, such as analyzing the corresponding x-ray signature of the specimen. However, scanning transmission electron microscopy techniques provide a broader perspective, as the transmitted electrons carry additional information for each scan position. This interplay becomes particularly valuable dealing with diverse measurement conditions, including specimen thickness and orientation, providing an opportunity to enhance the quantifiability of composition mappings. Importantly, this methodology transcends the limitations of individual techniques, leveraging the improved signal-to-noise ratio for thicker specimen in EDX over diffraction-based techniques, which are more effective with thin specimens. The resulting versatility not only enables precise composition determinations across a wider range of specimen parameters but also reveals additional synergies. For instance, it aids in resolving the indium concentration degeneracy in a pure analysis of the local form factor for InGaAs quantum devices. These insights not only amplify the quantifiability of S/TEM-based composition mappings but also harmonize well with novel approaches of composition determination, such as the analysis of stress relaxation for thin TEM-lamellas.

Graphic:



Keywords:

InGaAs, composition-mapping, Alloy-distribution, STEM-EDX, 4D-STEM

Reference:

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