Through-Focal HAADF-STEM of buried nanostructures

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Abstract. High-Angle Annular Dark-Field Scanning Transmission Electron Microscopy (HAADF-STEM) in combination with strain mapping techniques provides a powerful tool for quantitative analysis of crystalline semiconductor materials. Due to the complex interaction of a focused probe and a sample in HAADF, the calculation of each pixel in a simulation process requires a complete multislice iteration, making the overall computing process a rather demanding task in time and memory. SICSTEM is a parallel software code recently developed for running on the University of Cadiz Supercomputer (3.75 Tflops) that allows the simulation of images from large nanostructures containing more than one million atoms. The software has been designed to be able to generate not only one dimensional line scans or two dimensional images, but also to perform optical sectioning in the STEM simulation process, providing an easy way to simulate 3D HAADF-STEM images. In this work we consider GaAs capped GaSb nanostructures epitaxially oriented on a GaAs substrate. A methodology has been developed by combining the through-focal series STEM imaging and image analysis to estimate shape and position of buried GaSb nanostructures.

1. Introduction
The control of the shape, position and strain of nano-objects constituting self-assembled semiconductor nanostructures is a key aspect to improve their functionality. To improve the design of these nanostructures, it is important to have tools for quantitative interpretation. HAADF STEM image simulation tools can reveal information on individual atoms, but due to the high computational requirement needed, especially memory and time, nowadays can be only applied to a few unit cells. To improve these computational requirements, parallel HAADF STEM software has been developed. SICSTEM [1] is a HAADF STEM image simulation tool which is able to perform optical sectioning and to simulate aberration corrected microscopy, providing depth resolution at the nanometre scale in reasonable time.

The aim of this work is to estimate the 3D location of GaAs capped GaSb nanostructures epitaxially oriented on a GaAs substrate, from simulated images. Direct localization of buried nanostructures in three dimensions can be carried out by the intensity analysis of a through-focal series of images. A through-focal series is based on an acquiring process with the electron beam focused in a particular plane, then focus is updated to a new value by adding a fixed increment. Thus, the process consists of an iterative task of image acquisition and focus variation in order to get a set of frames from which information in 3D can be extracted.
Previous work demonstrated that the through-focal series acquisition provides the possibility of a 3D reconstruction of the sample investigated. In [2] it was shown that location of isolated heavy atoms in a matrix with a low atomic number was possible, although only third-order aberration correctors were used.

2. Experiment and results
In this work a square prism shaped GaSb quantum dot of dimensions 1×1×2 nm³ buried in GaAs (001) of dimensions 3×3×25.4 nm³ has been considered. The GaSb nanostructure was centred in the x-y plane, while in z direction three different locations were considered, namely at 4.24, 12.72 and 21.2 nm from the surface. Figure 1 shows the three models considered in this study where the position of each GaSb quantum dot can be clearly appreciated.

The corresponding supercells consisting of around 11800 atom coordinates, occupancy and Debye-Waller factors were created and applied as inputs to the SICSTEM software, using parameters for a 300 kV dedicated VG603 STEM ($C_s = -0.037$ mm, $C_5=100$mm, inner detector angle = 58 mrad, outer detector angle = 200 mrad and objective aperture = 22 mrad). In about 65 hours, a through-focal series containing 51 images per model of 512x512 pixels, with a focal step size of 0.5 nm was generated.

Each Z-contrast image along the <110> zone axis consists of atomic dumbbells formed by GaAs or GaSb atoms. Each dumbbell comprises two atomic columns, one containing Ga atoms throughout the whole material while each neighbouring atomic column is either pure As or pure Sb atoms. The intensity of Z-contrast images is strongly dependent on the atomic number of the elements that constitute each probed atomic column. Those peaks of a dumbbell with similar intensities correspond to GaAs ($Z_{Ga}=31$, $Z_{As}=33$), whereas higher intensities are more likely associated with Sb ($Z_{Sb}=51$).

We focus on the anion peaks where Ga atoms can be neglected. Figure 2 shows a profile taken at the centre of the image (inside the buried nanoprisim) and the other one near to the border (outside the buried nanoprisim). From a given simulated image the integrated intensities around anion peaks [3] have been calculated and the minimum value is assumed to be the As atom reference in the image.

![Figure 1. Three different models (1, 2 and 3) of GaSb quantum dot positions within a GaAs matrix.](image-url)
A significant relative difference between each integrated intensity and the reference value will determine the existence of Sb atoms, their \(x\) and \(y\) positions, and hence, the 2D shape of the buried nanostructure. Figure 3 (a) shows the determined shape at 22.5 nm depth for model 1 and (b) is a plot of normalised intensity differences for every anion position in the frame.

**Figure 2.** Simulated intensity profiles taken at the centre of an image (inside the buried nanoprism) and near to the border (outside).

**Figure 3.** Simulated image for model 1 acquired at 22.5 nm underfocus. The shape of the nanoprism is indicated by white full circles that represent Sb atoms; empty circles represent As atoms (left). Relative difference between each integrated intensity value and the reference value for every anion position marked in the frame (right).

In order to estimate the \(z\) position of Sb atoms, let’s define \(\Delta I_f\) as the relative difference between maximum \((I_{\text{max}})\) and minimum \((I_{\text{min}})\) anion integrated intensity at a given defocus value of \(f\), \(\frac{I_{\text{max}} - I_{\text{min}}}{I_{\text{min}}}\). This operation is carried out through the focal series of images. The highest value of the \(\Delta I_f\) set could be interpreted as the \(z\) position where the Sb atoms come into focus and are clearly visible.
Figures 4a, 4b and 4c show $\Delta I_f$ as function of the defocus values considered in the acquisition process for geometric models 1, 2 and 3. Maxima of these plots are found at around 4.5, 14 and 22.5 nm thickness, respectively, and in the corresponding geometric models these positions would correspond to the centres of the nanoprisms: 4.22, 12.5 and 21.2 nm. This discrepancy could be due to channeling effects. In order to determine the exact nanoprism length, new simulations considering different nanoprism sizes and positions will be necessary.

![Figure 4](image)

**Figure 4.** $\Delta I_f$ as function of thickness values considered in the acquisition process for geometric models 1, 2 and 3.

3. Conclusions
In this paper the shape and position of a GaSb nanoprism inside GaAs using HAADF-STEM focal series simulated images has been estimated. The proposed methodology can be applied to 3D reconstruction when nanostructures with heavy atoms are embedded in supercells with light atoms. Further research will be focused on applying this approach to experimental images. Problems may arise when segregation is present and/or due to the resolution in $z$-direction due to the channeling effect.

References

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