ANALYSIS of Dr.PROBE as a HAADF-STEM TOOL

James. Dalzell[1], Miryam. Arechavala-Arrendondo[1]
[1]*Centre for Nanostructured Media, Queen's University, Belfast, Northern Ireland.*

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Abstract— **The Dr.Probe simulation package was investigated in its ability to produce HAADF-STEM images of In doped GaAs, in which two methods of approach were evaluated. A method of producing doped material files through the manual creation of a doped superlattice was found to be more accurate than the single unit cell atomic occupancy approach, producing a Z power dependence of 2.228 compared to the theoretical range of 1.6 - 2.**

I. Introduction

High-angle annular dark field (HAADF) imaging is a scanning transmission electron microscope (STEM) technique that has been used extensively to obtain structural and compositional information of materials at the sub angstrom scale[1]. The technique produces a dark field image formed by electrons that have been incoherently scattered through very high angles (> 50mRads), where the intensity on the detector is largely determined by the Rutherford scattering process. This process is very sensitive to the atomic number (z) of the atoms within a target sample, due to its strong $Z^{(1.6-2)}$ dependence, and has consequently resulted in HAADF being a primary technique in producing Z-contrast images[2].

Due to the ease in which HAADF images can be understood, they have proven useful in supporting experimental results by providing the ability to: inspect grain boundaries, identify structural defects, and analyse interface fabrication quality upon viewing [2]. A key area in which HAADF-STEM has seen predominate use is in the study of semiconductor laser devices, in which noncommon atomic interfaces are continuously investigated to reduce any adverse effects originating from them influencing laser device performance [3]. One such effect results during the fabrication process in which differences in lattice parameters between two stacked material layers can produce a large amount of strain around a particular position, distorting the lattice at the interface producing structural defects. Dislocations caused by the high strain are highly detrimental to device performance, greatly affecting the material band structure by introducing non-radiative recombination centers. Reducing the origins of structural defects is paramount to improve performance, however many properties including the layer thickness can affect the strain distribution, which can prove time consuming and costly to explore. HAADF images can help in gathering qualitative information about the composition and site of potential defects but the process to gain fundamental quantitative insight, which could prove beneficial, is not a trivial task and requires the use of computer simulation.

Within this work HAADF-STEM images of In doped GaAs lattices were attempted to be examined under the Dr.Probe[4] software package. Dr.Probe has been shown to be a useful tool, capable of producing a range of high resolution STEM images across various modes, including HAADF. Since its development it has been successfully used in conjunction with experimental results to support measurements of atomic arrangements at interfaces, surfaces and defects, making it an interesting program to investigate further. The software functions through the application of the multislice method [5], an algorithm which calculates the forward scattering wave of a high energy electron following its interaction with a sample. Calculations are achieved using a FFT and preformed independently over all positions, with the fraction of the incident probe that falls within the specified angular range (>50 mrad) recorded and used to build up an image. The program has shown strong consistency with other simulation packages available but requires the careful inputting of experimental parameters and detector sensitives to function and provide accurate comparisons to experimental data. The goal of this work is to explore the capabilities of Dr.Probe in simulating the HAADF dependence of In doped GaAs under a range of percentages against sample thickness, while evaluating two methods of approach that can be used to produce the doped material files.

II. Methodology

GaAs ($P6_3mc$) typically viewed along the [110], was chosen for this study due to its mainstay in semiconductor laser design. For this study In atoms were doped, occupying Ga positions, for a range of concentrations and the effect on the HAADF intensity was measured as a function of sample thickness. The base GaAs lattice structure was displayed in the VESTA crystal making software, where two methods for doping the lattice structure could be followed. The first method focused on producing a superlattice of 3x3x5 unit cells along the [110] direction and replacing the Ga atomic positions, at random, with In atoms. The second method was to overlap Ga and In positions within a single unit cell and vary their occupancy rates depending on the required dopant level. In theory, when the same number of unit cells are simulated, the number of each atomic species in the sample should be equal, however this was not the case upon initial viewing in Dr.Probe with fractions of atomic species being produced along columns under the single cell method. With the goal to utilise Dr.Probe as a tool to support experimental results, both methods were to be explored to evaluate any differences which may potentially occur between them. For each method, the doped GaInAs file was simulated within Dr.Probe under an electron accelerating voltage of 200kV, a probe semi-angle of 22 mrads and an objective lens spherical aberration coefficient of 0.47 ± 0.01mm[6] ,given close to experimental values, with an annulus detector measuring 50-250mrads. Following the completion of each simulation, the HAADF images were imported to GATAN and the average intensity across the Ga/In columns was computed at 5nm intervals.

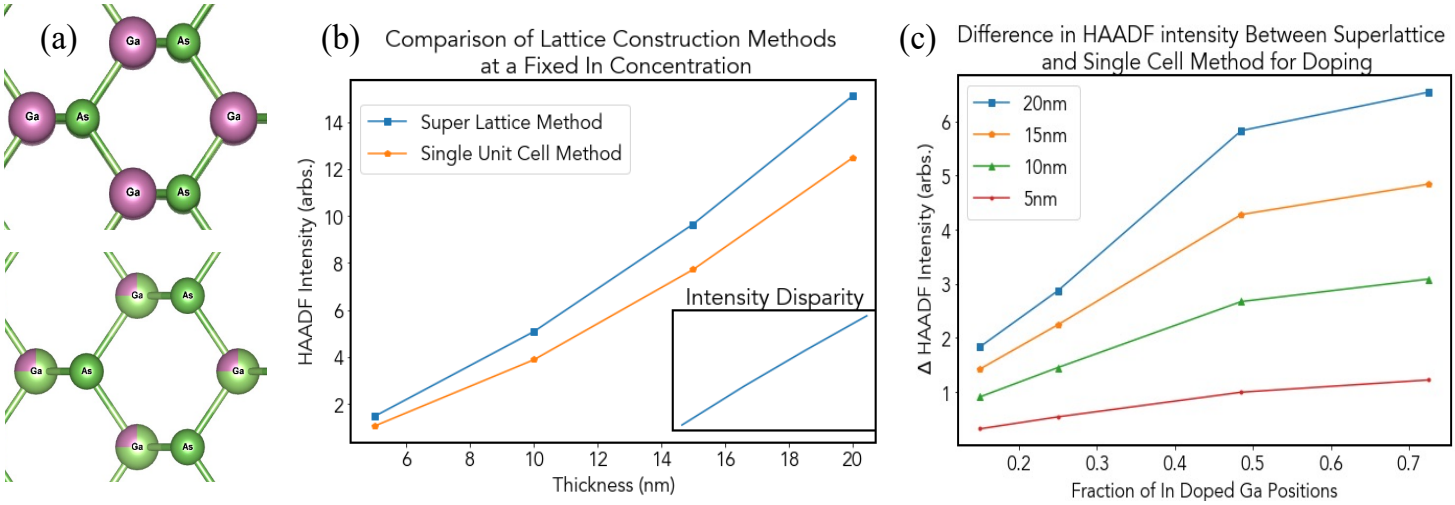

Fig 1: (a) Displaying a visualisation of the GaInAs [110] supercell (top) and single cell occupancy (bottom) approaches to doping within VESTA, with (b) and (c) comparing the HAADF results as a function of thickness for a fixed In concentration of 25% in (a) and a range of In concentrations in (b) (15%,25%,48.5% and 50%).

III. Results

GaInAs lattices were prepared within VESTA under both methods to contain 15%, 25% ,48.5% and 72.5% In atoms on Ga positions. All files were passed through the Dr.Probe software and modified to extend the crystal lattice to a maximum of 20nm. The multislice calculation was passed over the entire 3x3x5 lattice and repeated five times per scanning position, increasing computation quality, for a sampling of 10pm per pixel. The HAADF images were extracted and analysed within GATAN and the average intensity across all Ga(In) positions was calculated for thicknesses of 5,10,15 and 20nm before being compared to each other.

Figure 1b shows the dependence of the HAADF intensity on the sample thickness for a fixed 25% In concentration. The plot works as a viable basis to confirm that the simulation is functioning as intended due to both methods following a rough $Z^{1.6\text{-}2}$ dependence as more atoms are added down the column. It additionally proves that under a fixed doping concentration that there does exist a difference in intensity between the two methods. From the inset graph it can be seen that the difference between the superlattice and single cell methods is linear in nature under increasing thickness. To understand whether this is more a consequence of the linear behaviour of the multislice method with thickness, or the addition of more doped atoms as the thickness increases, an additional study was carried out focussing on the difference between the two methods as a function of In concentration and thickness.

Figure 1c displays a non-linear difference in HAADF intensity between the two methods as the dopant concentration increases, highlighting that the difference produced is largely a consequence of the two doping approaches. From this it can be theorised that the multislice method interprets/'sees' the doped species differently in the two approaches, with the origin perhaps lying in its development. Without approaching the multislice method on a theoretical level, the option was chosen to use a known relationship for HAADF to arrive at which method provided a more accurate representation of HAADF results. Comparing the doped columns intensity (I_1) to the undoped As columns (I_2), the following relationship could be followed[2] relating the intensities to each columns atomic number, where $\propto$ is typically found between 1.6 – 2,

$$\frac{I_1}{I_2} = \left(\frac{Z_1}{Z_2}\right)^{\propto}.$$

Taking the data for each dopant concentration and finding the value for $\propto$, the average for the superlattice method across all simulations was found to be 2.238, while a value of 1.115 was produced for the single cell approach. Both methods are outside the accepted range and more values under different dopants and lattice structures would need to be computed before an accurate conclusion can be made, but it could be suggested at this stage that for any future work taken that the manual superlattice approach should be followed.

IV. Conclusions and Future Work

It has been shown that the Dr.Probe tool could prove an excellent tool in simulating HAADF images, however there is some concerns over the best method to approach creating the required doped structure before interfaces of interest can be attempted to be simulated. Further work would be required to analyse both methods through a theoretical understanding of the multislice method and through empirical simulations within the software itself. If more work is to be done using the manual method, it should be suggested that producing a VASP file creation script could prove beneficial in reducing the time taken to produce the doped files. The HAADF intensity produced within Dr.Probe is also arbitrary in nature so additional approaches should be taken before comparisons can be made between the generated data and experimental results.

References

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