A HAADF Investigation of AlAs GaAs interfaces using SuperSTEM

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Abstract. Z-contrast imaging using a high-angle annular dark field detector can be used to characterise III-V heterostructures. GaAs/AlAs heterostructures were grown using MBE and prepared for TEM using a cross-sectional method. SuperSTEM 1 was used to investigate both the GaAs-on-AlAs and the AlAs-on-GaAs interfaces as a function of specimen thickness. The analysis of the images showed that the apparent interface widths varied with thickness in an unexpected manner. The GaAs-on-AlAs widths remained constant with thickness while the AlAs-on-GaAs interface widths increased. The actual interfacial width can be a result of either surface stepping during MBE growth or inter-diffusion of the column III atoms. To assist the interpretation of this result a series of interfacial models were created and explored using a frozen phonon multislice simulation. The models consisted of terraced, vicinal and diffused interfaces. The results show that while the width of the diffused interface reduces with specimen thickness, the widths of the stepped interfaces vary in a more complex manner. This variability is due in part to the different channeling depths of the Ga and Al columns, but the measured width is also affected by the depth of the material before the first step and whether GaAs or AlAs is on top. These effects can cause the apparent interfacial width to vary markedly from the actual width. Even measuring the width itself is non-trivial.

1. Introduction

The ability to control the growth of layers during molecular beam epitaxy (MBE) is vital for the development of sophisticated semiconductor devices. MBE techniques can currently produce very thin and abrupt layers with good control over parameters such as thickness, doping and composition [1]. With the advent of aberration correction it is now possible to study these interfaces at the atomic level using HAADF STEM imaging. HAADF imaging has been used at SuperSTEM to investigate the widths of [110] orientated GaAs/AlAs interfaces [2]. This paper reports multislice calculations used to aid interpretation of the experimental data.

2. Experimental Results

The experimental interfaces investigated were grown on a [001] orientated GaAs substrate by MBE. The samples consisted of a superlattice with repeats of 9 mono-layers of GaAs and AlAs. The samples were prepared using a standard cross-section method orientated in the [110] direction. The preparation method consisted of mechanical dimpling, ion milling and a final low kV polish. The experimental STEM images were recorded using SuperSTEM 1 with a convergence angle of 24 mrad at 100 kV giving a probe FWHM of ~0.1 nm. The HAADF collection angles were 70-210 mrad.

In the case of [110] orientated GaAs and AlAs the atomic columns align to form a series of dumbbells, see figure (1a), each dumbbell containing one As column and a type-3 column. For GaAs this type-3 column

3 SuperSTEM images courtesy of P D Robb.
would consist solely of Ga atoms while for AlAs it would consist of only Al. If the column is at the interface and has stepping or diffusion the column would contain a combination of both these atom types.

The HAADF detector collects the incoherent high angle scattered electrons. This scattering is Rutherford like and in the simplest approximation the scattering depends on the square of the atomic number, $Z^2$. The composition of a particular dumbbell can therefore be estimated from the column ratio. This is defined as the ratio of scattering from the type-3 column to that from the As column after background subtraction. Thus, in principle, HAADF images can be used to give an indication of the interfacial atomic structure. However the scattering of the STEM probe (and corresponding HAADF signal) depends greatly on the arrangement of atoms within the dumbbells. The electron density on an atomic column varies with depth, therefore the resulting HAADF signal will be sensitive to the depths of any compositional change. Because of this, it is necessary to compare experimental images to image simulations.

2.1. Column Ratio Profiling

The development of a consistent method for extracting column ratio data from HAADF images was developed [4]. This method was an attempt to standardise and automate the procedure of background removal, column ratio calculation and the subsequent interface width measurements. Figure (1a) shows the original HAADF image. Figure (1b) is a plot of the variation of the HAADF signal across a GaAs/AlAs interface from an averaged line profile from the region indicated in figure (1a). The solid trace is the HAADF signal, the dotted trace is the background and the dashed trace corresponds to the background subtracted dumbbell signal. The positions of the on column and background locations used in subsequent calculations are shown in figure (1c).

A background subtracted profile can be calculated for each row in the image, the dumbbell offset of each alternate row produces a set of interleaving line traces. Each alternate line trace is averaged together to form a complete column ratio profile. One such profile can be seen in figure (2a).

2.2. Width Measurement

The interface width is measured by least squares fitting an error function to the column ratio data, figure (2a). The width of the interface can then be defined as the distance between the 95% and 5% points on this curve. These values are bounded by the horizontal broken lines in figure (2a). This method can result in a width measurements of fractional values of a dumbbell, which can be interpreted as the averaged width in the [110] projection.

Figure (2b) is a plot of the experimental interfacial widths as a function of specimen thickness measured using this method [2]. ‘GaAs on AlAs’ corresponds to the interface where the GaAs layer was grown on an AlAs surface during MBE. Likewise ‘AlAs on GaAs’ corresponds to the interface where AlAs was grown on a GaAs surface. Figure (2b) indicates a clear asymmetry between these types of interfaces. The GaAs on AlAs interface widths are scattered around a constant width and appear to be independent of specimen thickness. However, the AlAs on GaAs interface widths increase with increasing specimen thickness.
Figure 2. a) Averaged column ratio profile with a fitted error function. b) plot of 5-95% widths as a function of thickness.

To develop a greater understanding of these results a series of interfacial models were developed that could describe this behavior. A simple diffusion model was formed where the interface width was due to intermixing of the type-3 atoms. A repeating terraced structure was also considered where the interface structure resembles a saw tooth with a characteristic step length. Another hypothesis was that the initial substrate was vicinal before commencement of MBE growth (the substrate polishing tolerance being ±0.5º). When the TEM sample is prepared and orientated this would cause the interfaces to also be vicinal and therefore the STEM probe positioned at the interface would project through one material before the other. These proposed interfaces were explored using a multislice code starting with an idealised perfect interface that can also be used as a control and a comparison for more complex structures.

3. Model Configuration
The interface model was constructed using a supercell with 14 x 12 x 300 unit cells (5.60 x 6.78 x 119.93 nm). The SuperSTEM probe was modeled including aberrations up to fifth order and a convergence angle of 24mrad giving a FWHM in agreement with the experimental probe. The simulations were calculated to thicknesses of 120nm and the effects of thermal diffuse scattering were included using the frozen phonon method, incoherently summing the results from 60 atomic configurations.

Figure 3. a) Plot of the interface width versus thickness for three models; b) Comparison of the experimental data to the best fit model.

4. Model Results and Discussion
Figure (3a) shows the results for three models of the ‘AlAs on GaAs’ interface. The diffusion model generally showed a narrowing as a function of thickness. The degree of narrowing was dependent on the rate of concentration change across the interface, with a gradual change in concentration giving the highest degree of narrowing. For the terraced interface the measured interfacial width was very sensitive to changes in specimen thickness below 60nm. For thicknesses above 60nm the sensitivity to further changes in column...
composition was greatly reduced. Neither model reproduced the experimental data. Thus it became apparent that a degree of vicinality was required to describe the experimental data.

The width of a vicinal interface inclined at $\theta$ to the normal increases linearly with specimen thickness. However the apparent width resulting from the above procedures is normally less than this and typically has a component that oscillates with thickness, see figure (3a). It also depends whether the probe is incident on GaAs or AlAs with the former case having no overall increase with thickness and the latter case increasing with thickness. This is the result of the channelling depth on the Ga column being $\sim$20nm whereas that on the Al is $>120$nm. The results are also very sensitive to the exact thickness of the layers involved.

While this is encouraging, it is not, in itself, enough to get good agreement. Figure (4) is the model which has given the best agreement to date. Here the value of $\theta$ is 0.4°.

However, at the GaAs on AlAs interface on the left, it is necessary to have some inter-diffusion (or terracing with a short step length) over a width of 2 dumbbells. At the AlAs on GaAs interface, it is necessary to have terracing with a random step length in the range 5–10nm [5]. The predicted profiles for this model are shown in figure (3b) along with the experimental data. The agreement is good but other models may prove to give equally good agreement. Thus the interpretation may not be unique. On the other hand, it is consistent with earlier work in which the GaAs on AlAs interface was described having microscopic roughness, while the AlAs on GaAs interface was described as atomically abrupt but stepped [5].

In conclusion, HAADF images of interfaces in which a large range of $Z$ is present need careful interpretation backed by appropriate modelling.

Acknowledgments
The authors would like to thank Martin Holland for growing the MBE materials, Brian Miller for specimen preparation and the superSTEM team. This work was funded by the Engineering and Physical Sciences Research Council research grant (GR/S41036) and DTA Studentship (M.F.).

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