Mg dopant distribution in an AlGaN/GaN p-type superlattice assessed using atom probe tomography, TEM and SIMS

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Abstract. P-type conducting layers are critical in GaN-based devices such as LEDs and laser diodes. Such layers are often produced by doping GaN with Mg, but the hole concentration can be enhanced using AlGaN/GaN p-type superlattices by exploiting the built-in polarisation fields. A Mg-doped AlGaN/GaN superlattice was studied using SIMS. Although the AlGaN and GaN were nominally doped to the same level, the SIMS data suggested a difference in doping density between the two materials. Atom probe tomography was then used to investigate the Mg distribution. The superlattice repeats were clearly visible, as expected and, in addition, significant Mg clustering was observed in both the GaN and AlGaN layers. There were many more Mg clusters in the AlGaN layers than the GaN layers, accounting for the difference in doping density suggested by SIMS. To evaluate the structural accuracy of the atom probe reconstruction, layer thicknesses from the atom probe were compared with STEM images. Finally, future work is proposed to investigate the Mg clusters in the TEM.

1. Introduction

GaN-based light emitting diodes (LEDs) are becoming ubiquitous as their use expands from niche applications towards everyday home and office lighting. These devices and others, such as laser diodes, require p-type and n-type layers for injection of the charge carriers that then recombine to produce light. Although n-type layers can be produced easily, often using Si as the shallow donor, p-type conductivity is much harder to achieve. Mg was the first dopant added to realise p-layers in GaN [1] and continues to be used in modern devices. The efficiency of such doping, however, is intrinsically low due to the deep acceptor state that Mg forms. By using an AlGaN/GaN p-type superlattice, the innate polarisation fields result in a
periodic oscillation of the valence band edge. This oscillation allows acceptors to be ionised wherever the acceptor level is below the Fermi energy, resulting in a hole concentration that is enhanced by up to 10 times.

A variety of characterisation techniques have been used to study a Mg-doped p-type superlattice. Although grown with nominally the same doping level in both the AlGaN and GaN layers, initial secondary ion mass spectrometry (SIMS) evaluation suggested that the AlGaN layers have a greater Mg content. Here, we present three dimensional (3D) atom probe tomography (APT) data to show the Mg distribution within the superlattice. The APT data are then compared with transmission electron microscopy (TEM) data to enhance the understanding of the superlattice structure.

2. Methods
The AlGaN/GaN sample was grown on a c-plane sapphire substrate using a 6x2” Thomas Swan close-coupled showerhead metal-organic vapour phase epitaxy (MOVPE) reactor. Trimethylgallium (TMG), trimethylaluminium (TMA), bis(cyclopentadienyl)magnesium (Cp₂Mg) and ammonia (NH₃) were used as precursors, with hydrogen (H₂) as the carrier gas. First, a ca. 30 nm GaN nucleation layer was deposited at 540 ºC, followed by a 2 µm GaN buffer layer grown at 1005 ºC. Next, the superlattice was grown at 1000 ºC with 24 repeat periods of ca. 10.5 nm Al₀.₁₁Ga₀.₈₉N and ca. 10.5 nm GaN. The Cp₂Mg flux during superlattice growth was 0.364 µmol/minute. The sample was then annealed at 780 ºC in N₂ for 1200 s. For the purpose of atom probe sample preparation, a non-intentionally doped 500 nm capping layer of GaN was added at 1005 ºC.

To study the Mg content in the layers of the superlattice, SIMS was conducted at Loughborough Surface Analysis Ltd. using a Cameca IMS 3f. Standard samples for Mg in AlGaN were not available at the time of testing, therefore only relative changes in the level of Mg can be reliably obtained from these data.

For the APT study of 3D Mg distribution, needle-shaped samples were prepared in a dual focused ion beam (FIB)/scanning electron microscope (SEM) by the standard lift-out and annular milling method described in Miller et al. [2]. An FEI Company™ Nova NanoLab™ with an Omniprobe Autoprobe™ 200 micromanipulator was employed. To remove areas of the sample damaged by ion beam milling at 30 kV, a low energy clean-up mill at 5 kV was used as the final preparation step. It has been shown that this minimises the ion beam induced damage to the APT sample [3].

The superlattice was then studied by APT on an Imago Scientific Instruments Corp. LEAP 3000X Si™. Under high vacuum and at low temperature (25 K), the sample was held at a standing voltage close to the threshold needed for field evaporation of the ions at the sample tip. A laser was focused onto the tip and pulsed at 200 kHz to induce field evaporation. Ion position and time-of-flight data were analysed using the Imago IVAS™ software. Once compositional identities were assigned to mass spectrum ranges and a number of other reconstruction parameters were defined, a 3D atomic map was produced.

Further structural characterisation was carried out using scanning TEM with a high-angle annular dark field detector (STEM-HAADF) on an FEI Tecnai F20-G2 with a 200 kV field emission gun. Sample preparation for the TEM foil was via standard mechanical polishing, followed by ion milling to electron transparency with a Gatan PIPS™.

3. Results and Discussion
Figure 1 shows the SIMS data for Mg and Al in the superlattice. The thick line is the Al concentration and the thin line is the Mg concentration. Although nominally doped to the same level in both the AlGaN and GaN layers, the Mg content appears to vary layer-to-layer. The dotted line in figure 1 illustrates that the AlGaN layers seem to have greater Mg contents. Although the Mg fluctuations appear to decrease with time, this is partially due to intermixing between layers as the primary ion beam penetrates further into the sample.
Figure 1. SIMS data for the superlattice with Mg doping to nominally the same level throughout. The thick line shows the Al and the thin line shows the Mg. There appears to be greater Mg content where there is more Al, corresponding to the AlGaN layers.

One of the strengths of APT is its ability to detect low concentrations of light elements such as Mg. Here, this ability is particularly relevant, since the Mg content of the superlattice is approximately 0.01 atomic % (at%); however, as shown in figure 2, it is not uniform. Ten percent of the Al ions collected are shown (dots) to indicate the AlGaN layers and a 4 at% isoconcentration surface is shown around the Mg ions. The areas of higher Mg concentration can be clearly seen and these will be referred to as ‘clusters’. To investigate the difference in Mg content between the AlGaN and GaN layers, the cluster density in each type of layer was analysed. There are many more clusters in the AlGaN layers, with 14.7 clusters on average in the AlGaN layers as compared to 1.7 clusters on average in the GaN layers (per layer volume of ca. 124,000 nm$^3$). Outside these clusters, the Mg content did not vary significantly between the two types of layers, therefore we conclude that the difference in Mg content suggested by SIMS is due to the greater density of clusters in the AlGaN.

To evaluate the structural accuracy of our reconstruction, the layer thicknesses measured from APT were compared with TEM. STEM-HAADF was used to image the superlattice, as shown in figure 3. The lighter contrast is GaN and the darker contrast is AlGaN. The layers were very regular, with an average thickness of $9.5 \pm 0.5$ nm for AlGaN and $10.7 \pm 0.2$ nm for GaN. The APT data for AlGaN layer thickness agreed well with the STEM.
giving an average thickness of 10.9 ± 0.2 nm, indicating the APT reconstruction parameters were accurate for this material. The GaN layer thicknesses were more inaccurately reconstructed in the APT, with an average thickness of 6.0 ± 0.2 nm, as currently released software allows only one global reconstruction field. It is important to note that if the reconstruction parameters were changed to alter the GaN thickness, this would not change the placement of the Mg clusters, as their positions would be similarly altered.

Figure 3. STEM-HAADF image showing the entire superlattice. The lighter contrast is GaN and the darker contrast is AlGaN.

Further TEM-based characterisation is planned to investigate the nature of the Mg clusters. Mg-rich, pyramidal defects in p-type GaN have been reported in the literature [4], always with the point in the [000-1] direction; however some controversy exists as to whether these are hollow voids [5] or inversion domains [4]. Future TEM on our superlattice may provide the opportunity to compare these defects with the clusters observed in APT.

4. Conclusions
SIMS data indicated that a Mg-doped AlGaN/GaN superlattice had greater Mg incorporation in the AlGaN layers. A 3D APT reconstruction of the superlattice showed Mg clusters, which were found to be preferentially located in the AlGaN layers. Outside the clusters there was approximately the same Mg concentration in both types of layers. The greater density of clusters in the AlGaN layers therefore accounts for the difference in Mg concentration observed by SIMS. STEM-HAADF was then used to measure the layer thicknesses in the superlattice. These values were compared to those found by APT, showing that the AlGaN layers were reconstructed more accurately than the GaN layers.

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