The impact of ScO$_x$N$_y$ interlayers on unintentional doping and threading dislocations in GaN

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Abstract. To reduce the threading dislocation density in (0001) GaN grown on c-plane sapphire, a series of samples have been grown using scandium oxynitride (ScO$_x$N$_y$) interlayers (ILs) on AlN-on-sapphire templates. Scanning capacitance microscopy (SCM) has been employed to investigate the unintentional doping in GaN with varying ScO$_x$N$_y$ IL thicknesses. The use of ScO$_x$N$_y$ ILs decreases the threading dislocation density. An unintentionally n-doped layer has been identified by SCM close to the GaN/ScO$_x$N$_y$ interface. The average width of this conductive layer has been quantified and found to increase as the ScO$_x$N$_y$ IL thickness increases up to 13 nm.

1. Introduction
Gallium nitride epilayers grown on c-plane sapphire often exhibit unintentional n-type conductivity with carrier densities in excess of $10^{16}$ cm$^{-3}$. Depth profiling Hall measurements suggested that this conductivity arises from a region adjacent to the GaN/sapphire interface [1]. Oxygen impurities have been found to be responsible for this conductivity [2], which may originate either from incorporation of impurities from precursor gases during growth [3] or diffusion from the sapphire substrate [4]. For GaN-based high power, high frequency electronic devices, and for optoelectronic devices whose functionality depends on the location and concentration of carriers, we need to both reduce the threading dislocation density and control or eliminate unintentional doping.

Identification and quantification of unintentionally doped regions in GaN epilayers has been recognized as an effective application of scanning capacitance microscopy (SCM) [3], as it measures carrier concentration and type separately. In this report we present an investigation employing SCM to examine unintentional doping in GaN samples in which scandium oxynitride (ScO$_x$N$_y$) interlayers (ILs) have been used to reduce the threading dislocation density (TDD). Previous studies have demonstrated the effectiveness of ScN ILs in TDD reduction. TDDs as low as $3 \times 10^7$ cm$^{-2}$ have been achieved [5]. In that case, faceted voids are generated in the GaN/sapphire template by GaN decomposition during the Sc IL nitridation and the TDD reduction mechanism may have involved dislocation bending during regrowth of the voided region, as well as dislocation blocking at the IL [6]. Here, by using AlN/sapphire templates and a partially oxidised Sc IL, we aimed to prevent void formation and investigate whether TDD reduction is still as effective. The effect of ScO$_x$N$_y$ ILs on both TDD and unintentional doping are described in this paper.
2. Experimental

Thin layers (5-25 nm) of Sc$_2$O$_3$ were deposited by magnetron sputtering of a Sc target in an oxygen/argon atmosphere on 1 µm AlN-on-sapphire templates grown by metalorganic vapour phase epitaxy (MOVPE). (The nominal Sc$_2$O$_3$ thickness was determined through interpolation of the deposition rate [7]). Then these layers were annealed in a 6×2 in. Thomas Swan close-coupled showerhead MOVPE reactor using 3 slm NH$_3$ and 17 slm H$_2$ at 200 Torr and 1070 °C for 30 minutes to form stable metal oxynitride. Detailed experimental procedures are given elsewhere [8]. Subsequent GaN was grown on the annealed ScO$_x$N$_y$ covered AlN-on-sapphire template. Three-dimensional island growth was promoted by using a low V/III ratio, and then these islands were coalesced using an increased V/III ratio and temperature. In-situ SiH$_4$ treatment at 860 °C was then carried out in order to highlight threading dislocation pits on the surface [9]; TDDs were then measured using an atomic force microscopy (AFM) in intermittent contact mode.

SCM measurements were carried out in cross section using a Veeco Dimension 3100 AFM mounted with a commercial SCM module. Cross-sectional sample preparation was achieved by cleaving, following the approach described in reference [10]. SCM was operated at a bias frequency of 90 kHz in the open loop mode and the AC voltages applied were in the range of 0-20 V peak to peak. PtIr-coated etched silicon tips with a nominal tip radius of 20 nm and a metallic layer thickness of 20 nm were used throughout this investigation. Topographic and SCM data have been obtained simultaneously during scanning across the sample’s cross-section. The fast scan direction was perpendicular to the AlN/sapphire interface. An automated routine was then carried out to extract the width of the unintentionally doped region in SCM dC/dV phase images as described in reference [2]. For each sample, at least 10 images with 256×256 pixels were taken, and the average width of the conductive layer was obtained. Image processing and data analysis were carried out using WSxM freeware [11].

3. Results and discussion

The threading dislocation density data for samples with initial Sc$_2$O$_3$ IL thicknesses from 0-20 nm are shown in Fig. 1. By using Sc$_2$O$_3$ ILs, TDD has been reduced from $9 \times 10^8$ cm$^{-2}$ to $1.8 \times 10^8$ cm$^{-2}$, showing the effectiveness of the Sc$_2$O$_3$ ILs in TDD reduction. The inset graph in Fig. 1 shows that TDD reduction becomes less significant as IL thickness is increased from 5 to 20 nm.

Figure 2 shows montages of SCM phase images of samples with nominal Sc$_2$O$_3$ IL thickness from 5-20 nm. All the images are displayed at the same scale. A dark layer with variable thickness is observed for all samples, which suggests that the GaN immediately above the ScO$_x$N$_y$ interlayer is n-type. (The position of the interlayer was identified from cleavage steps in topographic images, which are not shown here). The AlN and sapphire below and the GaN above the dark region appear grey and noisy, indicating that the carrier density in these regions is below the detection limit of the system.

In Fig. 3, the average width of the conductive layer is plotted against the initial Sc$_2$O$_3$ IL thickness. In the analysis, the regions affected by tip changes due to the cleavage steps perpendicular to the growth direction were excluded. However, other factors will also influence the accuracy of this width measurement, including the broadening due to the finite tip size used in SCM, carrier spillage and statistical errors. Figure 3 shows that the average width of the conductive interface layer increases with increasing IL thickness up to 13 nm. Assuming the lower surface of the conductive interface layer to be flat, the root mean square (RMS) roughness of the top surface of the conductive layer was...
calculated. This RMS roughness is also plotted with Sc$_2$O$_3$ IL thickness, showing a trend similar to the conductive layer thickness. It is noticeable that the width of the conductive interface layer first increases and then levels off with increasing Sc$_2$O$_3$ thickness, mirroring the behaviour of the TDD.

**Fig. 2.** Montages of SCM $dC/dV$ phase data for samples with Sc$_2$O$_3$ layer thickness from 5-20 nm.

Electron microscopy studies were carried out to check the ScO$_x$N$_y$ IL thicknesses for two of the samples and clarify the TDD reduction mechanism. Figure 4 shows cross-sectional transmission electron microscopy (TEM) images of the samples with nominal Sc$_2$O$_3$ IL thicknesses of 13 and 25 nm. The measured IL thicknesses from TEM are 17±0.8 and 19±0.5 nm, respectively. This discrepancy might partially explain the observed saturation in both TDD and conductive layer thickness. There is no evidence for the formation of voids in the AlN template for either sample.

It appears that the use of GaN/ScO$_x$N$_y$/AlN structure results in a less effective dislocation reduction in the overgrown GaN than for GaN/ScN/GaN samples. The TDD measured by AFM saturates at around $2 \times 10^6$ cm$^{-2}$, and similarly the SCM data suggest that the 3D-2D coalescence thickness does not vary for the interlayers with a nominal thickness of more than 13 nm. (For other samples, the width of the unintentionally n-doped layer has been found to increase as the coalescence time increases [2].) The absence of voids suggests that the mechanism of TDD reduction is different to that observed for GaN templates. The different mechanism may be due to the oxidation of the Sc or due to the thermal stability of the AlN seedlayer during the nitridation step which may result in the lack of pinholes formed in the interlayer. However, the observed saturation in both the TDD and the conductive layer thickness may not be an outcome of the growth mechanism but may have resulted from instabilities in the sputtering process resulting in inaccuracies in the thickness of the deposited Sc$_2$O$_3$.

The origin and mechanism of the unintentional doping in GaN are not fully understood. However, it has been suggested that most unintentional doping may be attributed to oxygen impurities [2], which in GaN growth on sapphire could come from contaminated precursors or carrier gases or from diffusion from the sapphire substrate. It has been found that oxygen incorporation is facet dependent and hence more oxygen incorporation may occur in the initial phases of GaN/sapphire growth where inclined facets are present [3]. Hence, the fact that an oxygen-doped region is often seen at GaN/sapphire interfaces does not necessarily imply that the oxygen comes from the sapphire. Inclined
facets can influence oxygen incorporation either from gaseous contaminants or diffusion. Despite the fact that the GaN films studied here were grown on AlN-on-sapphire templates with ScO\textsubscript{x}N\textsubscript{y} ILs rather than directly on sapphire, the ScO\textsubscript{x}N\textsubscript{y} interlayers might act as a source of oxygen, so we cannot state that in this case the unintentional doping definitely arise from gaseous contaminants. However, further studies of samples in which the oxygen content of the ScO\textsubscript{x}N\textsubscript{y} ILs is lower and gaseous contamination is different might provide new insights into this question.

4. Summary

SCM has been used to identify and quantify the unintentional doping in GaN grown on AlN-on-sapphire templates with ScO\textsubscript{x}N\textsubscript{y} ILs. The use of ScO\textsubscript{x}N\textsubscript{y} ILs decreases the TDD. An unintentionally n-doped layer has been identified close to the GaN/ScO\textsubscript{x}N\textsubscript{y} interface. The average width of this unintentionally doped layer has been quantified. TDD reduction using Sc\textsubscript{2}O\textsubscript{3} ILs and AlN templates appears to have a different mechanism to that seen with ScN\textsubscript{x} ILs and GaN templates. Most TDD reduction methods – including that studied here – appear to lead to increased unintentional doping.

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