Oxygen segregation to nanopipes in gallium nitride

M Hawkridge and D Chrens

H H Wills Physics Laboratory, Royal Fort, Tyndall Avenue, Bristol BS8 1TL

ABSTRACT: The formation of nanopipes in GaN has been linked to impurity segregation. In this paper, a combination of high angle annular dark field imaging and electron energy loss spectroscopy in the Daresbury SuperSTEM is used to investigate the core structure and composition of open core dislocations (nanopipes) in GaN films grown by hydride vapour phase epitaxy. The results show evidence for segregation of oxygen to the nanopipe surfaces. Quantitative analysis suggests that up to several monolayers of nitrogen can be replaced by oxygen. The implications of these results for understanding the electrical properties and core structure of dislocations in GaN are discussed.

1. INTRODUCTION

Threading dislocations in GaN are known to affect the optical and electronic properties of devices. In order to understand these properties, understanding of the core structure and composition of dislocations is required. In fact, recent work has suggested, albeit indirectly, that segregation of impurities to dislocations is a major factor affecting the core structure of dislocations.

Threading dislocations in GaN are of 3 types, with Burgers vectors 1/3<11-20> (edge dislocations), 1/3<11-23> (mixed dislocations) and <0001> (screw dislocations). Our previous work has shown that in undoped GaN grown by metal-organic chemical vapour decomposition (MOCVD) screw dislocations are of open core type (nanopipes) whereas edge and mixed dislocations appear to have closed cores (Cherns et al 1997). In MOCVD grown GaN heavily n-doped with Si (Cherns et al 2000), nanopipes were non-uniform containing constricted (closed core) segments as well as more open structures. In contrast, MOCVD Al$_{0.03}$Ga$_{0.97}$N heavily doped with Mg showed strong evidence for Mg-segregation to dislocations with both edge and mixed dislocations being open core on a fine scale (Cherns et al 2002). In GaN grown by molecular beam epitaxy (MBE) under Ga-rich conditions, we have reported open core dislocations of mixed type (Baines et al 2003). These open core dislocations were decorated with amorphous material believed to contain excess Ga, although no conclusive proof of this was obtained.

In this paper, we examine the core structure of dislocations in GaN grown by hydride vapour phase epitaxy (HVPE). A combination of high angle annular dark field (HAADF) imaging and electron energy loss spectroscopy (EELS) in the Daresbury SuperSTEM is used to show the presence of substantial quantities of oxygen on the surfaces of open core screw dislocations. The results are compared with transmission electron microscope (TEM) observations on plan-view and cross-sectional specimens. The significance of the results for understanding the core structure and the electronic properties of dislocations in GaN are discussed.

2. EXPERIMENTAL

A series of samples with varying thickness of epitaxial layer (0.6, 5 and 55μm) were grown by HVPE on (0001) oriented sapphire substrates. No buffer layer was employed to reduce dislocation density and no deliberate removal of impurity was made. Electron transparent samples were prepared in both plan view and cross-sectional orientation by a standard mechanical polishing technique and dimpling, followed by low-energy ion milling in a Gatan PIPS™ thinner to electron transparency.
Samples were examined in a Philips EM430 TEM operating at 250kV and in the 'SuperSTEM' at Daresbury Labs, UK.

The SuperSTEM is equipped with an HAADF detector and an electron energy loss (EEL) spectrometer that allows for high resolution lattice imaging in parallel with chemical analysis. The nature of HAADF and EELS also means that results are virtually directly interpretable, after some refinement.

After recording an atomic image, line scans were made across the cores of several nanopipes in each sample of varying GaN layer thickness. Each scan was selected to include the oxygen and nitrogen K-edges and the gallium M2,3-edge. For each point along the scan, a quantitative measure of each element’s concentration was extracted from the EELS data by fitting a power law to a 20-100eV window preceding the edge of interest to strip away the phonon background. An appropriate integration window (>20eV) was then selected after the edge from the background stripped data to give a number proportional to the areal density of the corresponding element. In order to compare the amount of each element to the others present, the integrated data was scaled to the N bulk signal. The O was scaled to the N by using scattering cross-sections and the Ga signal was scaled to the N by assuming that the average Ga signal was equal to the average N signal away from the dislocation core.

3. RESULTS AND DISCUSSION

![HAADF image of open dislocation core (nanopipe) in 0.6µm film, b) close-up of hexagonal atomic structure and c) composition profile corresponding to line of scan.](image)

Figure 1 shows a HAADF image of a nanopipe set in the hexagonal GaN bulk looking down the [0001] zone axis. The lattice image is proportional to $Z^2$ so that generally the brighter the image, the more material there is. The close-up of the HAADF image clearly shows the lattice structure of atoms as bright points on a dark background. HAADF is also advantaged with minimal coherent scattering effects to consider such as contrast oscillations related to thickness variations. The faces of the pipe lie on the {10-10} planes and the spatial resolution is about 1.3Å. Below the HAADF image, the refined EELS data is shown yielding the compositional variation across the scan. A Burgers circuit drawn on the HAADF images and cross-sectional TEM analysis showed that the nanopipes are screw type.

As the edge of the pipe is approached, the N signal is observed to drop away from the bulk concentration (marked on the lattice image by an unfilled arrow). This coincides with a rise in the O