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The structure and properties of dislocations in GaN

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Transmission electron microscopy studies of the core structure and optoelectronic properties of dislocations in GaN films are described. It is shown that the core structure depends sensitively on the growth method and on the presence of dopants and impurities including Si, Mg and O, with edge, screw and mixed dislocations all becoming open core type under certain conditions. High-resolution electron energy-loss spectroscopy is used to confirm impurity segregation to dislocations. Electron holography and cathodoluminescence studies showing that dislocations possess band gap states and act as non-radiative recombination centres are reviewed, and correlated, tentatively, to impurity segregation.

1. Introduction

GaN/InN/AlN films are now used in a range of light emitting diodes and laser diodes emitting at the blue end of the visible spectrum, and are expected to lead to new electronic devices for high-speed, high-power applications. It is, however, surprising that such devices work, given that the active regions contain high densities of threading defects, up to 10^9 cm^{-2}. These defects, mostly dislocations, arise from the current need to grow the GaN on highly mismatched substrates. In order to clarify the role of dislocations, a detailed understanding of their structure and properties is required. This paper describes our recent work in this area. We show how the core structure of dislocations in GaN can be open or closed, and depends on the dislocation type, the growth method, and on the presence of dopants and impurities. Studies of the optical and electronic properties of dislocations are then briefly reviewed. Finally, the significance of segregation, and of the core structure, in understanding these properties are briefly discussed.

2. Experimental

We have examined GaN films grown by metal-organic chemical vapour deposition (MOCVD), hydride vapour phase epitaxy (HVPE) and molecular beam epitaxy (MBE) under a range of doping conditions. In all cases, these films had the hexagonal wurtzite structure and were grown in the (0001) orientation, mostly on (0001) sapphire substrates. Transmission electron microscopy (TEM) studies were carried out on both plan-view and cross-sectional samples prepared by standard methods of mechanical polishing and Ar-ion thinning.

Studies were carried out on dislocations threading through the GaN layers. The dislocations were of the three types normally associated with hexagonal materials, namely a-type, with Burgers vectors $\mathbf{b} = 1/3\langle 11\bar{2}0\rangle$, $c + a$-type, with $\mathbf{b} = 1/3\langle 11\bar{2}3\rangle$ and c-type, with $\mathbf{b} = \langle 0001\rangle$. These three dislocation types are illustrated in Fig. 1. Since dislocations tend to follow the growth direction, normally the [0001] or c-direction (perpendicular to the basal (0001) plane) they are often referred to as edge dislocations (a-type), mixed dislocations ($c + a$-type) and screw dislocations (c-type).

3. Results

In general the GaN films studied contained a variety of defects. This is illustrated in Fig. 2, which shows a plan-view sample of undoped GaN grown by MOVCD. The sample has been tilted from the horizontal to show threading defects, which are mostly parallel to [0001], in projection. The defects include inversion domains (I), nanopipes (N), which are hollow tubes with diameters 10–30 nm across, as well as threading dislocations (D). Studies by large-angle convergent-beam electron diffraction (LACBED) have established that, in undoped GaN grown by MOCVD, nanopipes are, in fact, open-core screw

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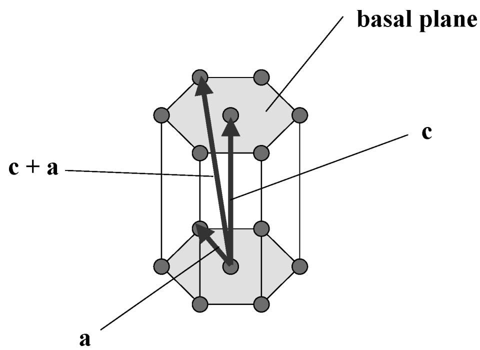


Figure 1 Schematic showing the Burgers vectors of the three types of dislocation observed in (0001) GaN films.

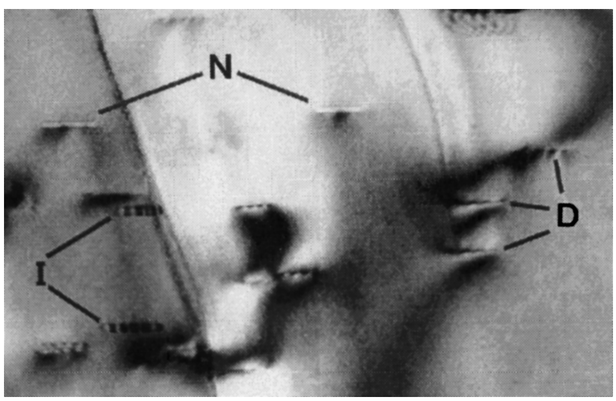


Figure 2 Threading defects in a plan-view sample of MOCVD grown GaN: I = inversion domains, N = nanopipes and D = undissociated dislocations.

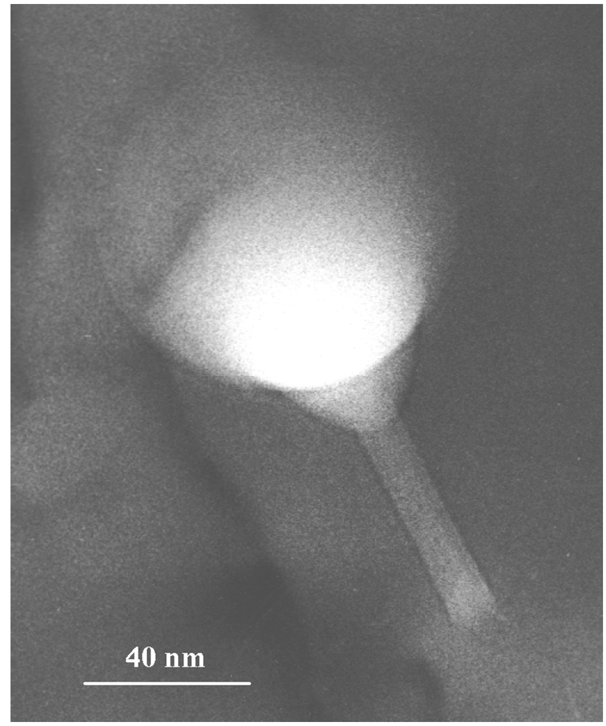


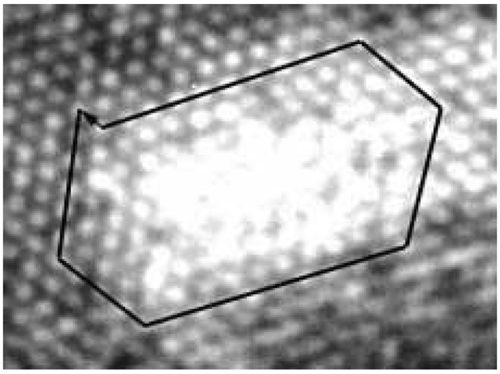
Figure 3 A nanopipe in Si-doped GaN, which opens out into a hollow cone.

dislocations [1]. In contrast, there is little evidence that edge and mixed dislocations in this material have open cores. Indeed, weak-beam and high-resolution lattice imaging suggest that these dislocations are also undissociated.

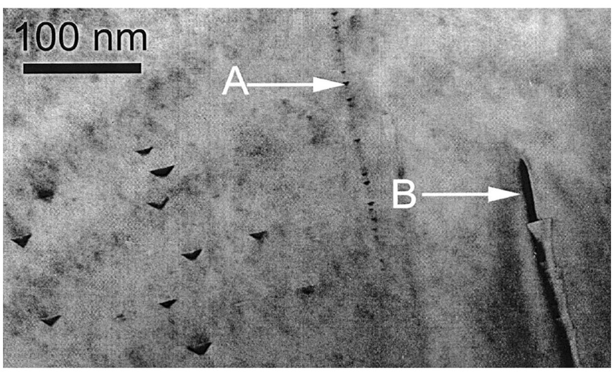
There has been a considerable debate about the origin of open-core dislocations which have now been observed in a number of hexagonal materials [2]. Frank [3] proposed that dislocations with large Burgers vectors might become open core to reduce total energy, i.e. by replacing strain energy with the energy of newly-created surfaces. However, for screw dislocations in GaN, which have a Burgers vector of 0.5 nm, the Frank theory suggests an equilibrium core radius of the order of an atomic spacing. Northrup [4] has shown that substantial reductions in the surface energy, which would lead to larger equilibrium radii, are possible if the core is decorated by Ga. Alternatively, nanopipes might form for kinetic reasons. For example, we have suggested instead that nanopipes could form from pinholes formed during the early stages of GaN growth [1]. Such pinholes would contain no surface steps if a trapped screw dislocation was present and therefore might propagate as a nanopipe if step-flow growth dominated. The situation is different for pinholes associated with edge and mixed dislocations where surface steps would be present, thus causing the nanopipe to grow out as growth proceeded.

It is now clear that the core structure of threading dislocations also depends on the growth method, and on the presence or absence of dopants. In MOCVD samples which were heavily n-doped with Si, we have observed open-core screw dislocations of varying core diameter. Some contained segments which were completely constricted, thus appearing as normal, closed core, screw dislocations, and others opened out into large hollow cones as illustrated in Fig. 3.

In contrast, observations on MOCVD-grown AlGaN films, which were heavily *p*-doped with Mg, showed that edge and mixed dislocations had hollow cores [5]. Some results are illustrated in Fig. 4. Fig. 4a shows a lattice image of a hollow-core dislocation in a plan-view sample seen in end-on orientation along [0001]. The dislocation is of edge or mixed type, as confirmed by the Burgers circuit which is superimposed. Fig. 4b shows edge and mixed dislocations, A and B respectively, in a cross-sectional sample. The diffracting conditions are such that the edge dislocation is out of contrast, but has its position marked by a high density of precipitates which decorate the dislocation core. Larger precipitates of the same morphology (hexagonal pyramids visible in projection as triangular defects) are visible in the bulk. Such precipitates are known to be associated with Mg segregation. More extensive observations showed that



(a)



(b)

Figure 4 Dislocations in Mg-doped $Al_{0.03}Ga_{0.97}N$ (a) plan-view sample: lattice image of an edge or mixed dislocation viewed end-on, (b) cross-sectional sample: showing decoration of edge and mixed dislocations.

regions close to edge and mixed dislocations were relatively denuded of precipitates. The mixed dislocation in Fig. 4b is also decorated, but in this case the decorating defects appear to have void-like character.

To explain the observations in Fig. 4, it was proposed that Mg, an oversized atom, was segregated to edge and mixed dislocations owing to the elastic interaction with the dilatational part of the stress field. This can account both for the absence of precipitates in regions close to the core of edge and mixed dislocations, and the decoration of the cores themselves. Although we have no conclusive proof of Mg segregation to dislocations (in the form of microanalytical data), we believe that, if segregation occurs, the Mg inhibits the growth of GaN locally leading to a pit. This, in turn, becomes a void if subsequently overgrown. A similar mechanism has been proposed to explain the presence of voided regions seen sometimes in Mg-related precipitates in bulk material. In this case it is supposed that growth is inhibited by the nucleation of a slow-growing inversion domain, which has been recently confirmed both by high-resolution and convergent-beam electron diffraction studies [6].

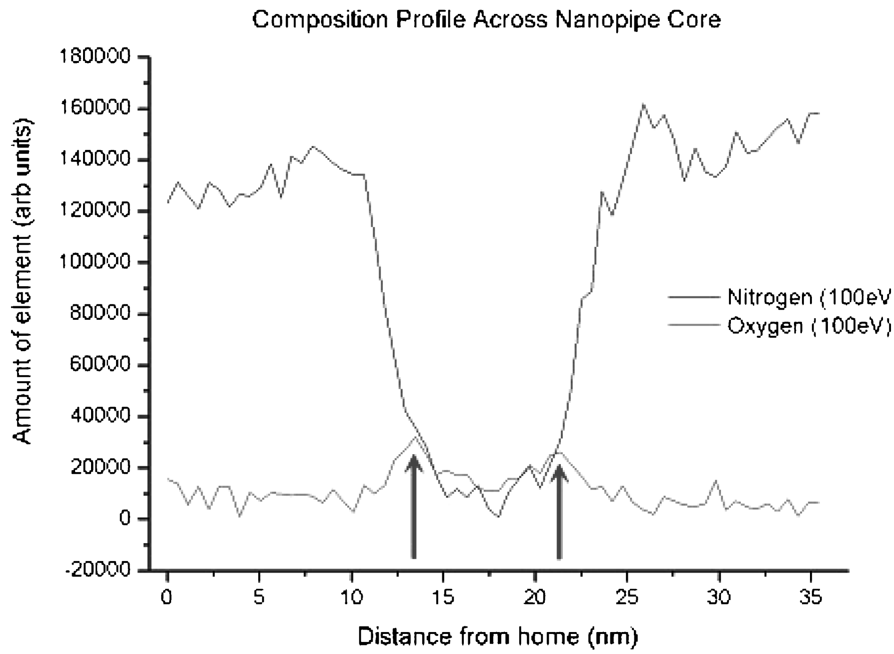
In GaN films grown by MBE under heavily Ga-rich conditions, we have also observed open-core dislocations but this time of mixed-type [7]. The observations suggest that the cores are decorated with an amorphous deposit, believed to contain excess Ga. Recent studies of a range of samples of this type grown under very similar conditions have suggested that open core formation varies from one sample to another and between different regions of the same sample. The reasons for this behaviour are not clear, although there appears to be a correlation between open core formation and the presence of associated surface pits [8]. This suggests that segregation of excess Ga to surface pits, which are often but not exclusively associated with threading dislocations, may be the key factor in the formation of Ga-decorated cores.

We have used high spatial resolution imaging and electron energy-loss spectroscopy (EELS) studies in the Daresbury SuperStem to clarify the structure and composition of open-core dislocations. Fig. 5 illustrates studies carried out on a nanopipe in HVPE-grown GaN. The high-angle annular dark field (HAADF) image shows a plan-view sample with a nanopipe viewed in end-on orientation along [0001]. The lattice image enabled us to construct a Burgers circuit around the nanopipe. This indicated no net displacement, consistent with the nanopipe being an open core screw dislocation. The approximate edges of the nanopipe are indicated by arrows on the corresponding EELS line profiles. These show nitrogen and oxygen profiles, oxygen being a likely contaminant in HVPE growth. The line profiles show the K-edge signals, which have been scaled by comparison of the theoretical cross sections to give relative atomic concentrations of N and O. The results suggest that within a few nanometers of the core there is evidence of replacement of N with O. More extensive studies suggested that some nanopipe cores (including that in Fig. 5) contained an amorphous deposit containing Ga and N.

Studies of cross-sectional samples of the HVPE layers confirmed the presence of open core screw dislocations, but showed that some were non-uniform along their length, containing pyramidal voids not unlike those seen in Mg-doped material. An example is shown in Fig. 6. These results suggest that the edges of open core defects may not be vertical in Fig. 5, suggesting that the broadening of the oxygen peak over several nanometres may be a projection problem rather than indicating that the oxygen concentration falls off gradually from the surface. Further work is needed to clarify this point.

4. Optoelectronic properties of dislocations

We have developed methods to examine the optical and electronic properties of individual dislocations as a function of their Burgers vector. To examine the optical properties, we have used low-temperature cathodoluminescence studies in the scanning electron microscope (SEM-CL). In GaN, the use of low-incident-beam energies (1–5 keV) results in high spatial resolution, since beam spreading is small (less than 0.1 μm) and the generated carriers



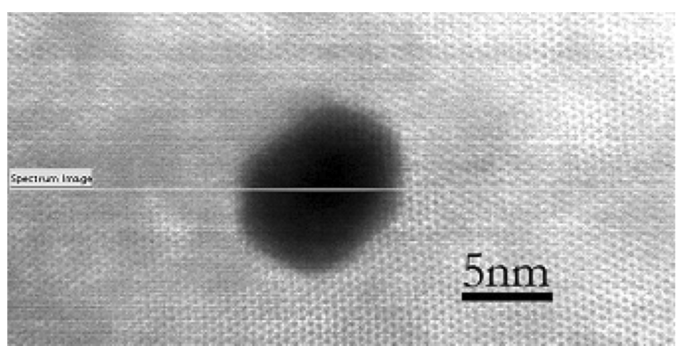


Figure 5 A screw dislocation in HVPE-grown GaN seen end-on in the Daresbury SuperStem in a HAADF image, and corresponding EELS line profiles for N_K and O_K signals.

generally have relatively short migration distances before recombination takes place (less than 0.2 μm). In addition, it has proved possible to generate CL in samples which have been prepared for transmission electron microscopy (TEM), thus allowing us to correlate CL maps with later TEM studies on the same samples. In studies of GaN/InGaN samples with thin (2 nm) InGaN quantum wells and thin capping layers, we have thus been able to show that individual edge dislocations act as non-radiative recombination centres for InGaN band edge luminescence [9].

We have examined the electronic properties of dislocations using off-axis electron holography in the transmission electron microscope (TEM-EH). Interference fringes in an electron hologram are displaced depending on the relative phase difference between a beam passing through an object and that of a reference beam. The phase difference at low spatial resolution is given by $\delta\varphi = CV_0t$, where C is a constant, V_0 is the mean inner potential (assumed constant throughout the film thickness) and t is the sample thickness. By examining dislocations in an end-on configuration, we have been able to profile changes in the inner potential as a function of distance from the core. Some results are illustrated in Fig. 7. Fig. 7a shows a hologram taken from an edge dislocation in MOCVD-grown GaN, which is n-doped with Si [10]. The dislocation has been tilted by a few degrees from the exact end-on orientation, [0001], to minimise changes in diffraction contrast due to the dislocation strain field. Such effects can affect the local mean potential sampled by the beam owing to channelling. The dislocation is just visible through residual black/white contrast where the dislocation intersects the top and bottom of the foil. The experimental hologram was then converted to a phase map. Fig. 7b shows the calculated potential derived from the phase map, indicating that the potential at the dislocation core is about 2.5 V different from that in the bulk.

Figure 6 A screw dislocation in HVPE-grown GaN in a cross-sectional sample showing decoration by voids.

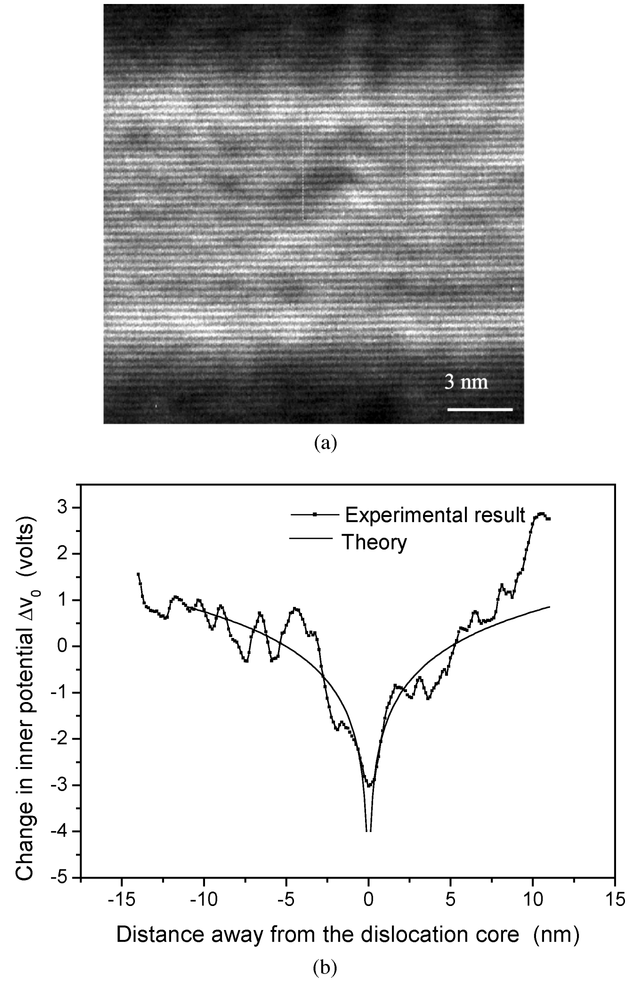


Figure 7 (a) Electron hologram of a near end-on edge dislocation in n-doped GaN. The dislocation shows some residual contrast where the ends intersect the foil surfaces (in the outlined box), (b) a plot of inner potential derived from the phase map.

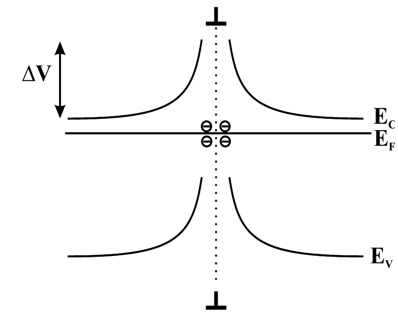


Figure 8 Model of band bending to explain the results in Fig. 7.

The TEM-EH results in Fig. 7 can be explained by the presence of a negative charge on the dislocation core, which is equivalent to about $2e/c$, where $c = 0.5$ nm is the unit cell dimension along the c-direction ([0001]). Studies of screw and mixed dislocations in n-doped GaN show results which are similar to those for edge dislocations, although, in this case, the contribution of changes in specimen thickness is uncertain [11]. In contrast, TEM-EH studies of p-GaN (Mg) showed quite different results with the inner potential increasing near the core of some dislocations [12]. These results indicate a positive charge on dislocations in p-GaN although the magnitude is lower than observed in n-GaN (around 0.6 V). In bandstructure terms, the charging of dislocations observed by TEM-EH can be explained in a model of band bending, induced by pinning of the Fermi level at states in the GaN band gap. This is illustrated in Fig. 8 for the case of n-type material. To agree with the results in Fig. 7, the pinning level should be around 2.5 V below the conduction band edge. A similar model can be derived for p-type GaN, with band bending up to 0.6 V in the opposite direction, implying states close to those derived for n-GaN.

5. Discussion

The TEM-EH studies demonstrate that dislocations of all types in GaN should be highly charged, as a result of Fermi level pinning at states relatively deep in the band gap. Calculations suggest that edge and screw dislocations in stoichiometric material should have no band gap states, although the conclusions depend on whether the cores are of open or closed type [12]. This suggests that the band gap states should be of extrinsic rather than intrinsic origin. This has also been confirmed by calculations indicating that a range of point defects, including Ga vacancies, and vacancy complexes with impurities such as oxygen, can give rise to band gap states [13, 14].

The TEM results described here confirm that segregation of impurities to the cores of dislocations in GaN is a major factor. These may be intentional impurities, such as Mg (Fig. 4), or unintentional impurities such as O (Figs 5 and 6). It is also clear that, in addition to providing a source of extrinsic states, impurity segregation is sufficient to cause dislocations to become open core. The fact that edge, screw and mixed dislocations can become open core under appropriate growth conditions suggests that kinetic rather than energy factors are of primary importance, although further work is clearly needed to confirm this point.

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