HOMO- AND HETERO-EPITAXIAL GALLIUM NITRIDE GROWN BY MOLECULAR BEAM EPITAXY

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Abstract

Various methods have been used to initiate growth by Molecular Beam Epitaxy (MBE) of GaN on sapphire, or other substrates, but there is always a problem with morphology and with a high defect density which results in the formation of a sub-grain boundary structure. We show that by using, homo-epitaxial growth on properly prepared bulk GaN substrates, combined with high temperature growth, we obtain a significant improvement in surface morphology. Growth at sufficiently high temperature leads to a rapid smoothing of the surface and to almost atomically flat surfaces over relatively large areas. Multi-Quantum Well structures grown on such GaN epitaxial films are dislocation free with abrupt interfaces.

Introduction

The Group III-Nitrides are an important class of semiconductors now being used for light emitting diodes (LEDs), short wavelength blue/ultra-violet (UV) laser diodes (LDs) and high temperature electronic devices. LEDs are already commercially available from a number of suppliers [1,2]. Recently, blue/UV laser diodes have been demonstrated with room temperature cw operational lifetimes in excess of 3000 hours [3]. Projected lifetimes of >10000 hours have been announced at a number of recent international conferences. Other device structures based on the group III-Nitride system, grown both by Metal Organic Vapour Phase Epitaxy (MOVPE) and Molecular Beam Epitaxy (MBE), including high-power high-frequency FETs [4] and solar blind UV photo-detectors [5] have also been reported.

During the growth of GaN layers, either by MOVPE or MBE, one would ideally grow under stoichiometric conditions. However, for films grown by MBE, it is difficult to achieve growth under exact stoichiometric conditions and films are usually grown slightly Ga or N-rich. Films grown under N-rich conditions show a columnar structure [6] and they are not suitable for device purposes. Films grown under Ga-rich conditions at low temperatures show large hexagonal features of wurtzite GaN, in a polycrystalline background and with additional Ga droplets. It is common practise, therefore, to use a slightly Ga-rich growth mode at high temperature, the excess Ga being desorbed.

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Amongst the various growth methods for group III-nitrides, MBE has provided most insight into the growth kinetics, because of the in-situ analytical measurements such as reflection high energy electron diffraction (RHEED). A variety of GaN RHEED patterns have been seen and reported in the literature, during or after MBE growth, these include (1×1), (2×1), (2×2), (2×3), (3×2), (3×3), (4×6) and (5×5). The un-reconstructed (1×1) pattern has been shown to correspond to a monolayer of Ga, which is tightly bound to the GaN [7,8]. On top of this relatively stable Ga adlayer, there are mobile Ga adatoms which give rise to the surface reconstruction.

One of the most severe problems hindering both MBE and MOVPE progress in this field is the lack of a suitable substrate material on which lattice-matched group III-nitrides films can be grown. So far the best device results have been obtained on insulating sapphire or SiC substrates. For most optoelectronic devices a vertical configuration is normal; however, growth on dielectric substrates such as sapphire, requires a planar device configuration, which is a disadvantage for many of the present GaN-based devices. Growth on sapphire and SiC substrates leads to a large lattice mismatch and a significant difference in thermal expansion coefficient, which in turn leads to high defect density in the epitaxial wafers. Unlike the other III-V compounds in which defects such as dislocations are detrimental to device performance, the nitride films are relatively unaffected. For example, the presence of a high density of dislocations does not appear to limit the efficiency of LEDs [9], but more recent evidence suggests that this is because of the low diffusion rate for carriers in GaN [10]. There is also evidence that dislocation motion in nitride semiconductors is much lower than in other III-V compounds [11]. However, it has been suggested that the electron mobility in GaN depends strongly on the dislocation density [12]. Therefore, bulk GaN substrates would be the ideal choice for a number of reasons; there would be zero lattice and thermal mismatch, a conducting or insulating substrate would be available, the dislocation density in epitaxial films would be low, the thermal conductivity would be high and the cleavage planes would be perpendicular to the (0001) surface.

The purpose of this paper is to demonstrate that by using high quality bulk GaN substrates and appropriate MBE growth conditions, dislocation free atomically flat GaN epitaxial films and Multi-Quantum Well (MQW) structures can be obtained. By contrast films grown under equivalent conditions on sapphire have subgrain boundaries and inferior properties.

Experimental Details

The high quality bulk GaN substrates used in this study were grown from the Ga solutions at nitrogen pressures of 12-15 kbar and at temperatures of 1500-1600°C. They were hexagonal platelets 6-8 mm in size, highly conductive if grown from the solutions in Ga or semi-insulating if doped with Mg during growth. Due to the absence of inversion symmetry, the GaN substrate has two distinctly different crystal orientations - (0001) and $(000\bar{1})$ corresponding to the Ga and N polarity respectively. The $(000\bar{1})$ hexagonal face corresponding to the N-polarity is chemically active and therefore it could be prepared for epitaxy by mechano-chemical polishing [13], which gives atomically flat surfaces without subsurface damage. All the studies reported in our paper were obtained on this polarity.

The MBE growth was performed in two different systems, a commercial Varian Modular GEN-II system equipped with a high density RF source (HD25) from Oxford Applied Research (OAR) and a purpose built mini-MBE system equipped with a CARS25 RF source also from OAR. The GEN-II system has elemental Al, Ga, In, Si and Mg sources and the mini-MBE system an elemental Ga source in addition to the nitrogen source. Growth rates in the two systems are typically 0.8 and 0.3 μm/hour respectively under stoichiometric conditions. More details of the growth procedures are reported elsewhere [14]. The substrates were mounted on molybdenum holders with indium, with gallium-tin alloy [15], or on a special designed metal-free holder. RHEED patterns prior

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to, during and after growth were monitored using a reflection high energy electron diffraction equipment. In the GEN-II system a video camera is linked to a high quality Super VCR system and in the mini-system a video capture card is used to store images on a PC system.

The Atomic Force Microscope (AFM) images of the grown layers and structures were obtained using a Topometrix Explorer 2000 system non-contact mode.

Results and Discussions

Typically for both MBE and MOCVD, GaN growth on sapphire is implemented as a three step process [16]. For MBE, the substrate is first exposed to a nitrogen plasma [17] to change the surface to AlN [18]. Next a GaN or AlN buffer layer is grown either at low temperature, 550°C [17], or at higher temperatures, 750°C [19]. Finally epitaxial layers are grown at about, 750-850°C. The aim of this three step process is first to wet the sapphire surface with GaN or AlN and to minimise the effect of the large lattice mismatch between substrate and epilayer. Nevertheless, the region close to the interface is structurally poor, with a very high dislocation density.

In our experience, GaN films grown by MBE on sapphire, even under optimum conditions, show rather poor RHEED patterns compared with other III-V compounds. Growth at a low substrate temperature, 600°C, produces a high density of small, oriented sub-grains. Growth at higher temperature, about 750-850°C, results in a similar structure, but on a much larger scale, as illustrated in Figure 1. In this case, the individual sub-grains are typically 0.3 to 0.5 μm diameter and a detailed AFM study of one of the individual islands shows that they are nearly atomically flat with a root mean squared (rms) roughness of 0.1 nm in the best cases. Such films are continuous with typical electron mobilities of 180-200 $cm^2V^{-1}s^{-1}$ at a carrier density of around $5x10^{17}$ cm^{-3}.

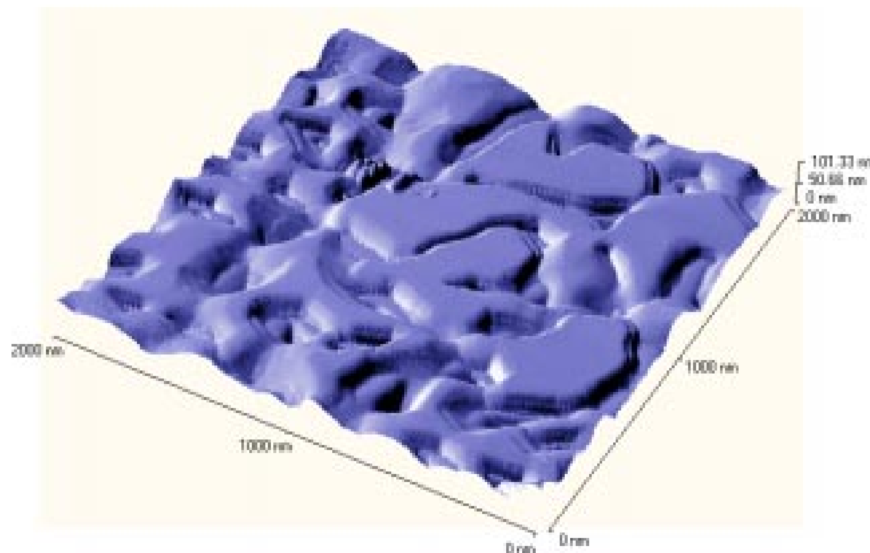

Figure 1. 3D AFM image of GaN layer on sapphire substrate.

For example, (InGa)N/GaN MQWs grown on such GaN on sapphire substrates using this MBE method, show very poor structural and optical properties. Transmission Electron Microscopy (TEM) studies of such MQW structures demonstrate that we have non-planar QWs, which follow the non-planar heterointerface between the GaN epitaxial layer and the QWs. The optical properties of the MQWs, show rather weak broad emission with no band-edge related features.

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By contrast, growth on bulk GaN substrates show completely different behaviour. We have grown GaN layers on the bulk GaN crystals at temperatures >750°C, under slightly Ga-rich conditions. During growth, we usually observe a streaky (1 × 1) RHEED pattern, which becomes stronger after the first minutes of epitaxy. Sometimes, during growth we can see a (2 × 2) RHEED pattern. Strong Kikuchi features are observed and the intensity of the specular spot is much stronger than in films grown on sapphire. On cooling the GaN layers grown under slightly Ga rich conditions, a clear (2 × 2) reconstruction is always observed below ~300°C.

There are two different configurations for Ga on the surface which can possibly give rise to a (2 × 2) reconstruction. The first arises from additional Ga on top of the first complete Ga monolayer, this (2 × 2) reconstruction is formed on cooling to low temperature via an order/disorder transition from excess Ga on the surface. The second (2 × 2) configuration can potentially arise from vacancies in the first layer of Ga, which terminates the surface, this (2 × 2) reconstruction is formed by evaporation of a finite number of Ga atoms from the Ga terminating layer. It is entirely reasonable that increasing the temperature will increase the probability of desorption for Ga and that, therefore, a vacancy induced (2 × 2) reconstruction is entirely possible especially during growth at high temperatures. Further studies are in progress to clarify this situation.

In contrast to growth on sapphire, AFM studies of GaN surfaces grown by home-epitaxy, show remarkably flat surfaces over large areas as shown in Figure 2. Line scans in the non-contact mode show atomically flat surfaces over large area with overall rms roughness for a typical 2x2 μm^2 surface of <0.1 nm over the whole area. It is important to note that there is about two orders of magnitude difference between the scales in Figure 1 and Figure 2. We can conclude therefore that the growth proceeds in the 2D mode where the lateral growth rate is very much larger than the growth rate normal to the direction of growth. TEM studies of the GaN films grown under such conditions show no additional dislocations in the epitaxial layer, whereas similar studies on GaN films grown on sapphire show dislocation densities of $>10^9$ cm^{-2}, even under optimum conditions. In addition, the high optical quality of the homo-epitaxy of GaN has already been demonstrated [20,21].

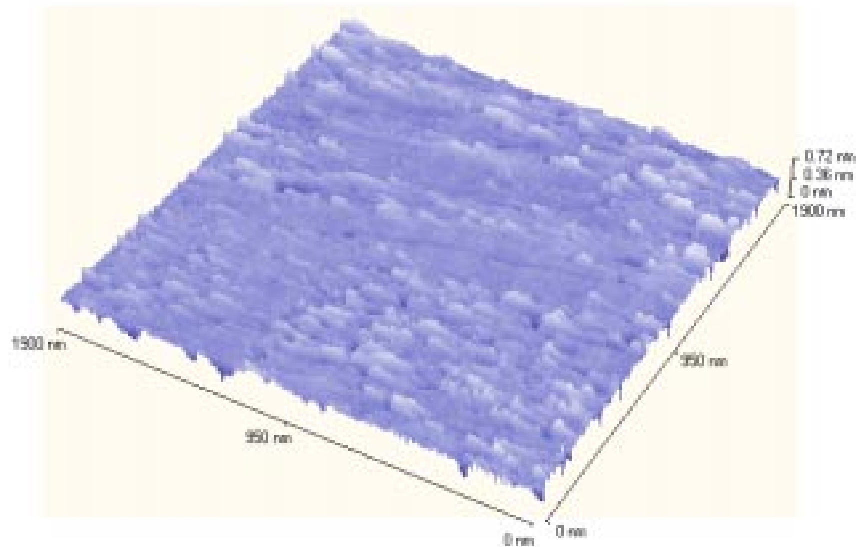

Figure 2. 3D AFM image of GaN layer on bulk GaN substrate

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We have also grown (AlGa)N/GaN MQWs on top of our homo-epitaxial GaN layers. The growth rates were similar to those used for the GaN and the growth temperature was unchanged. TEM studies of such structures show that, in contrast to MQWs grown on sapphire substrates, high quality dislocation free structures are observed for large areas (>250nm), as shown in Figure 3. In other regions we see some evidence for wavy interfaces, but the origin of the perturbation is not yet established. In this figure almost atomically abrupt interfaces are observed between the QW and barrier regions. The above films have also been characterised using X-ray diffraction. Powder diffractometer studies show additional satellite peaks from the MQW periodicity and the experimental data follows closely the theoretical model for the ideal structure.

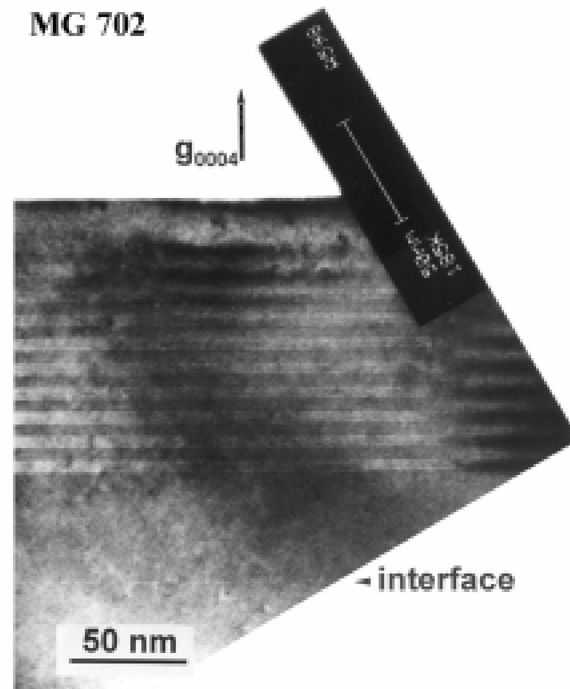

Figure 3. TEM image of an AlGaN/GaN MQWs grown on bulk GaN substrate.

Conclusions

Whilst various techniques for the initiation of growth on sapphire, or other substrates with a large mismatch, can improve the quality of films grown by MBE, there remains a fundamental problem with morphology. The high density of dislocations results in the formation of a sub-grain boundary structure. By contrast, homo-epitaxial growth under appropriate conditions on properly prepared bulk GaN substrates results in greatly improved morphology and RHEED patterns. Growth at sufficiently high temperature leads to rapid smoothing of the surface and to atomically flat surfaces over relatively large areas. Multi-Quantum Well structures grown on such GaN epitaxial films are dislocation free with very abrupt interfaces.

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