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Defect reduction with quantum dots in GaN grown on sapphire substrates by molecular beam epitaxy

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The GaN films grown on buffer layers containing quantum dots by molecular beam epitaxy on sapphire substrates were investigated. The density of the dislocations in the films was determined by wet chemical etching and atomic force microscopy. It was found that the insertion of a set of multiple GaN quantum-dot layers in the buffer layer effectively reduces the density of the dislocations in the epitaxial layers. As compared to the dislocation density of $\sim 10^{10}$ cm^{-2} in the typical GaN films grown on AlN buffer layer, a density of $\sim 3\times10^{7}$ cm^{-2} was demonstrated in the GaN films grown with quantum dot layers. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1432445]

III-nitride semiconductors have wide applications in light emitting, detecting, and electronic devices and have been investigated very extensively in the last decade.[1,2] GaN based light emitting devices (green, blue, violet) and laser (violet) diodes have been achieved using heterostructures and quantum wells.[3] III-nitride heterostructures are commonly grown on foreign substrates by metalorganic chemical vapor deposition (MOCVD) and molecular beam epitaxy (MBE).[4] Sapphire (α-Al_2O_3) substrates are most extensively used owing to their relatively low cost and large size. Reasonably high quality GaN has been grown on sapphire and quality improves with lateral epitaxial overgrowth (LEO). However, the large mismatches of lattice constant, thermal expansion coefficient, and stacking order of III-nitrides from substrates result in high defect density in the epilayers. Although the density of certain dislocations near the GaN surface may decrease with the film thickness, the typical value for a GaN layers with thicknesses in the range of few to ten μm grown on sapphire substrate is still in the order of 10^{9} cm^{-2} or higher.[5] This value is too large as compared to $\sim 10^{4}$ cm^{-2} in homoepitaxial GaAs films and certainly affects the electrical and optical properties as well as the device performance.

Attempts have been made to grow GaN films with reduced defect density. Thick GaN layers have been prepared by techniques such as hydride vapor phase epitaxy (HVPE) and used as the substrates (templates) for further growth by MBE and MOCVD to reduce defects. Using GaN platelets grown by the high-pressure technique, the dislocation density as low as 10^{5} cm^{-2} has been reported in the GaN/AlGaN quantum well structures grown by MBE.[6] The photoluminescence (PL) efficiency was greatly improved as compared to the heteroepitaxial materials.[7] Using GaN templates grown on sapphire by HVPE, an electron mobility higher than 50 000 cm^2/Vs was observed in an AlGaN/GaN two dimensional system with an electron density of 2.8×10^{12} cm^{-2}.[8] In addition to HVPE prepared substrates, GaN templates grown by MOCVD on sapphire via lateral epitaxial overgrowth have also been used as the substrate for MBE re-growth. A room temperature electron mobility of about 1200 cm^2/Vs was measured in a GaN film grown on such templates.[9] The dislocation density of $\sim 5\times10^{8}$ cm^{-2} was reported.

Since the native substrates are not available in sufficient size and quantity, alternative methods by using various types of buffer layers for improved nucleation and reduced defect density have been investigated. In the case of MOCVD, a thin GaN or AlN film grown at low temperature (~500 °C) is commonly used as buffer layer for the active layer growth which takes place at much higher temperatures. Other buffer layers such as low temperature InN layer on sapphire *A*-face,[10] double low temperature AlN or GaN layers inserted between high temperature grown GaN,[11,12] GaN/AlGaN superlattices,[13] and the SiO_2 patterned GaN/AlN buffer on SiC substrate (LEO or pendio epitaxy)[14] were all investigated in an effort to reduce defects. Recently, the effects of the growth rate,[15] the thermal annealing,[16] and the impurity doping[17,18] of buffer layers were also reported. As compared to MOCVD growth, which also could employ LEO growth on patterned substrates,[19] and insertion of multiple low temperature GaN buffer layers,[11,12,20] much fewer choices, if any, are available for MBE. Each of these approaches has its own advantages and disadvantages are considered a stopgap measure, and LEO requires patterning and overgrowth with the associated Si autodoping. In the case of MBE, an AlN buffer layer is commonly used as the buffer layer and a high growth temperature (~900 °C) followed by the deposition of GaN layers at moderate temperatures.[21,22] The nitridation temperature of sapphire substrate[23] and the III/V flux ratio[24] were found to have an impact on the GaN quality.

In this paper, we report sizeable reduction in defect density, as studied mainly by defect delineating etches, in the GaN quality by using multiple layers of quantum dots (QDs) in the buffer layer. Utilizing defect delineating chemical etching (which has been verified to be reliable on control

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samples) on the GaN films grown with and without GaN/AlN QDs, we demonstrated that the etch pit density can be reduced from $\sim 10^{10}$ cm^{-2} to $\sim 10^{7}$ cm^{-2} by employing quantum dots.

Three samples reported in this manuscript were grown on *c*-plane sapphire substrates by MBE with both ammonia and radio frequency (rf) activated nitrogen sources. The growth sequence of the sample A (sample #743) include an AlN layer (~30 nm) on substrate, a GaN layer (~400 nm), a second AlN layer (~10 nm), a second GaN layer (~300 nm), and a third AlN layer (~10 nm). All three AlN layers were grown at high temperature of ~950 °C. Although a low temperature AlN may lead to a smoother surface, but x-ray diffraction (XRD) and post-GaN growth experiments indicated that the high temperature buffer layers result in better crystal quality. The GaN layers were grown at ~800 °C with a growth rate of ~0.5 μm/h.

In addition to the above structure, a 20 period GaN/AlN QDs was grown in two additional growth experiments. The growth temperature for the GaN/AlN QDs was ~800 °C. The GaN layers were about 4 atomic monolayers thick (~1 nm in thickness). The AlN spacer layers were about ~2 nm thick. For the second sample B (sample #734), the growth was terminated after the 20 periods GaN/AlN QDs was completed. For the third sample C (sample #744), an additional GaN layer (~300 nm) was grown at a temperature ~750 °C on the top of QDs. The details of QD preparation and resultant optical properties have been reported elsewhere and will not be repeated here.[25,26]

The samples were characterized by XRD and PL measurements. The widths of the symmetric (002) peaks in the XRD spectra are 1.1 arc-minute for sample B and about 2 arc-minutes for sample A and C. The asymmetric (104) peaks are broader (~8 arc-minutes). The PL spectra from all three samples show band edge luminescence at 3.4 eV at room temperature. The efficiency of the PL from sample B and C is, however, much higher than that from the sample A. From samples B and C, the luminescence bands from GaN/AlN QDs were also observed.

The density of dislocations in the epilayers was examined by the wet chemical etch and atomic force microscopy (AFM). Hot (160 °C) phosphoric acid (H_3PO_4) was used as the chemical etchant. It has been demonstrated that this etchant only attacks the defect sites in Ga-polar (0001) GaN surface and that etch pit density determined as such is comparable to the dislocation density determined by TEM, Photo Electric chemical etching, and KOH etching in control samples.[27,28] The pits, mostly in hexagonal shape, appear after the surface is etched. To reiterate, the pit density measured by AFM has been correlated to the dislocation density in the film and the validity of the etching process for defect delineation has been established previously for HVPE samples by comparing to TEM investigations.[27,28]

A typical AFM image of the as grown and the etched surface for the sample A is shown in Figs. 1(a) and 1(b), respectively. The vertical scale is 30 nm for image 1(a) and 20 nm for 1(b). The size of both images is 1 μm$\times$1 μm. The etching time for the image 1(b) is 15 s. The result shows the high density of defects. A very rough estimate of the dislocation density by accounting the number of the etched

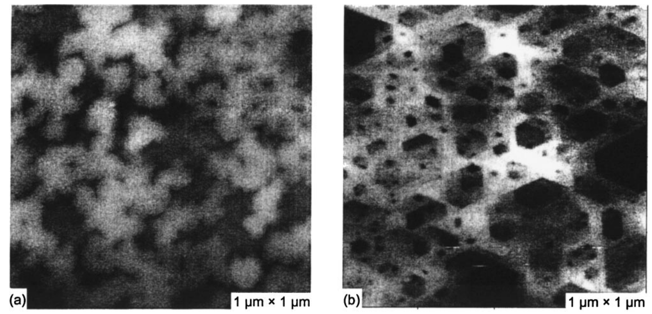

FIG. 1. AFM images of sample A (sample #743). (a) As-grown surface, vertical scale is 30 nm (b) Etched at 160 °C H_3PO_4 for 15 s, vertical scale is 20 nm.

pits is on the order of 10^{10} cm^{-2}. This is a typical value for the GaN films grown by MBE on sapphire substrates using AlN as the buffer layer, which are inherently thin.

An AFM image of the as grown and the etched surface for the sample B is shown in Figs. 2(a) and 2(b), respectively. The vertical scale is 20 nm for image 2(a) and 30 nm for 2(b). The size is 1 μm$\times$1 μm for image 2(a) but 10 μm$\times$10 μm for 2(b). The enlarged scan from the etched surface does not show additional pits. The etching time for the image 2(b) is 5 min. The surface morphology of the sample with different etching time was also examined. It was found that the change in the etching time from 2 to 15 min only affects the pit size but not the density. The pit-free areas were also hardly affected by the etching time. The well separated etch pits allows us to reliably count the pit density, which is about 3×10^{7} cm^{-2}. As demonstrated in the HVPE grown GaN, these pits represent the etched dislocations in the GaN samples and the pit density gives the dislocation density near the sample surface.[27,28] Thus, as compared to sample A, a reduction in the dislocation density by nearly three orders of magnitudes in the sample B is evident.

For sample C, the AFM image of the as grown and the etched surface is shown in Fig. 3(a) and 3(b), respectively. The vertical scale is 30 nm for image 3(a) and 50 nm for 3(b). The size is also 1 μm$\times$1 μm for image 3(a) but 10 μm$\times$10 μm for 3(b). The etching time for image 3(b) is 12 mins. Similar to the sample B, the change in the etching time in this case again only affects the pit size but not the density. The pit density, $\sim 4\times10^{7}$ cm^{-2}, was also close to that observed in sample B, demonstrating a low dislocation density in this film.

We believe that the significant reduction in the disloca-

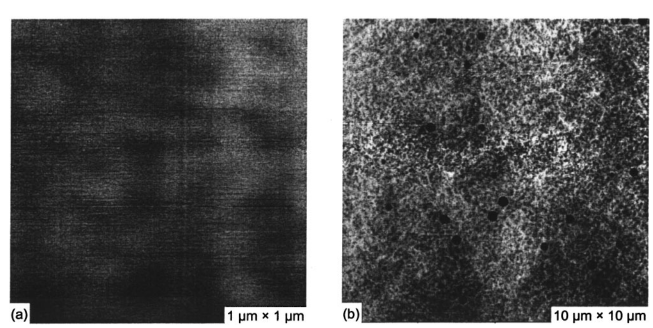

FIG. 2. AFM images of sample B (sample #734). (a) As-grown surface, vertical scale is 20 nm. (b) Etched at 160 °C H_3PO_4 for 5 min, vertical scale is 30 nm.

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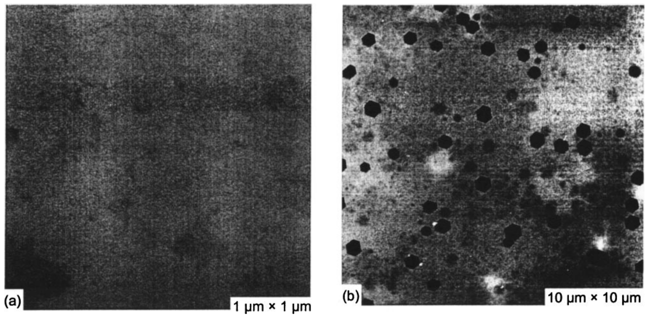

FIG. 3. AFM images of sample C (sample #744). (a) As-grown surface, vertical scale is 30 nm. (b) Etched at 160 °C H_3PO_4 for 12 min, vertical scale is 50 nm.

tion density revealed by the etch pits in the samples B and C, is essentially due to the introduction of the multiple QDs layers. In the case of lateral growth on the SiO_2 patterned GaN surface,[14] it was found that the dislocations from the GaN were blocked by the SiO_2 and the lateral growth on the SiO_2 shows a very low dislocation density. In our case, the dislocations extending from the film/substrate interface in the samples B and C appear to be interrupted by GaN QDs. For example, the dislocation lines that have an in-plane component may loop around the QDs and no longer extend into the sample surface. Dislocations may also terminate at the surface of the dots. The partially lateral growth of the GaN QDs and the AlN spacer layer may also have contribution to the observed low dislocation density. The detailed mechanism of the dislocation interaction with GaN/AlN QD layers is not yet well understood at this time and is a subject of further investigation.

In conclusion, we have investigated the crystalline quality of MBE grown GaN films containing quantum dot defect filters on sapphire substrates by defect delineating chemical etching. A pit density, correlated to dislocation density, of $\sim 3\times 10^{7}$ cm^{-2} was observed in the GaN films grown on the multiple GaN/AlN QD layers as the buffer. This value represents a reduction in the dislocation density of nearly three orders of magnitudes as compared to the films without using any QD layers.

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