Growth of crack-free high-quality GaN on Si(111) using a low-temperature AlN interlayer: observation of tilted domain structures in the AlN interlayer

Min-Ho Kim, Young-Gu Do, Hyon Chol Kang, Chel-Jong Choi, Do Young Noh, Tae-Yeon Seong, and Seong-Ju Park*

Department of Materials Science and Engineering, Kwangju Institute of Science and Technology, Kwangju 500-712, Korea

Received 4 April 2003, revised 20 June 2003, accepted 22 August 2003
Published online 20 November 2003

PACS 61.10.Nz, 78.66.Li, 81.15.Gh

We report on the growth of crack-free high-quality GaN layers on Si(111) using a low-temperature AlN interlayer (LT-AlN IL) between GaN epilayers by ultrahigh vacuum chemical vapor deposition. The use of a thin LT-AlN IL resulted in the complete elimination of cracks and significant improvements in structural and optical properties of the GaN layer. The GaN epilayer containing the LT-AlN IL exhibited a strong band-edge emission with the line width of 26.5 meV at room-temperature. Additional distinct domains with a tilt angle of ±0.9° in the LT-AlN IL, found by synchrotron X-ray diffraction, are believed to play a key role in the effective relaxation of thermal stress and the reduction of threading dislocations.

© 2003 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

1 Introduction The growth of III-nitride semiconductors on Si substrates is an important technology in achieving novel optoelectronic integrated circuits, which consist of InGaN/GaN optical devices and Si-based microelectronics. However, the large differences in intrinsic material properties between GaN and Si(111) substrates such as the large degree of lattice mismatch (~17%), large differences in the thermal expansion coefficients (~37%), and their polar/nonpolar properties, give rise to the generation of extensive defects and, in particular, to the formation of dense microcracks along {1100} in GaN layers. It has been reported that cracks act as nonradiative and scattering centers in light propagation and electron transport, thus resisting in-plane electrical current, and producing shortcuts in vertical currents [1]. Therefore, it should be noted that a reduction in the density of cracks is needed to achieve acceptable device performance and reliability. In the case of the growth of GaN on sapphire substrates, a variety of types of interlayers between GaN epilayers have been employed to achieve these goals and have been reported to successfully reduce dislocations and/or cracks in (Al)GaN layers [2, 3]. However, the detailed structure of the low-temperature interlayer has not yet been clarified. In particular, only a few reports have appeared on the effects of the low-temperature AlN interlayer (LT-AlN IL) on the growth of GaN on Si(111) substrates [4, 5]. In this paper, we report on the growth of crack-free GaN layers via the use of LT-AlN IL between GaN epilayers on Si(111) grown by an ultrahigh vacuum chemical vapor deposition (UHVCVD) system. In addition, we also report on the effects of tilted domain structures in the LT-AlN IL on the elimination of cracks and significant improvements in the structural and optical properties of GaN layers.
Experiment

The samples were grown on Si(111) substrates using triethylgallium (TEGa), trimethylaluminium (TMAI), and NH₃ (99.9999% purity) in a UHVCVD reactor, as described in previous publications [6, 7]. An AlN/AlGaN double-layered buffer (DLB) was used for the growth of GaN. The AlN layer was grown at 830 °C without any prior surface nitridation process in order to avoid the formation of a silicon nitride phase on the silicon surface, followed by the deposition of the AlGaN layer for a DLB and a 1 µm-thick GaN epilayer at 800 °C. The temperature was then slowly ramped down to ~550 °C for the growth of a thin AlN interlayer (~30 nm). The temperature was then increased to 800 °C to grow a 1 µm-thick GaN overlayer. For a comparison study, two samples (sample A: GaN (2 µm)/DLB (AlGaN (60 nm)/AlN (20 nm)/Si(111), sample B: GaN (1 µm)/LT-AlN IL/GaN (1 µm)/DLB (AlGaN (60 nm)/AlN (20 nm)/Si(111) were grown under the same growth conditions. To examine structures of GaN and an LT-AlN IL, high-resolution synchrotron X-ray scattering measurements were carried out at the 5C2 beamline at the Pohang Light Source (PLS) in Korea. Intense synchrotron X-rays in a high-resolution configuration made it possible to investigate structures of a thin LT-AlN IL as well as the in-plane structures of GaN epilayers. We measured the conventional X-ray diffraction profiles (θ-2θ scan and θ-rocking) along the substrate-normal Q_z direction. Perfect in-plane GaN(10T0) reflections under a grazing indicant geometry were also measured to study in-plane structures near the surface of the GaN layers. The Q_z-component in the reciprocal space was maintained at Q_z = 0.06 Å⁻¹, and, as a result, their penetration depth was less than 3000 Å. PL measurements were also performed at room temperature using a He–Cd laser (325 nm) with an excitation power of 15 mW.

Results and discussion

Room-temperature PL spectra for samples A and B are shown in Fig. 1. In both samples, the band-edge emission of the wurtzite GaN epilayer was predominant without the defect-related yellow-band emission near 2.3 eV. Compared to GaN without the LT-AlN IL (sample A), sample B exhibited sharper band-edge emission with full width at half maximum (FWHM) of 26.5 meV. The PL intensity also increased by an order of magnitude. This indicates that the use of the LT-AlN IL is effective in improving the optical properties of GaN. The FWHM of 26.5 meV from sample B is smaller than the best reported value for GaN on Si(111) substrates [6–8]. In addition, it should be noted that samples A and B show slight different peak positions of the band-edge emission. The band-edge emission of sample B appeared at a slightly different energy side than that of sample A (ΔE~5 meV). This shows that GaN grown with the LT-AlN IL is under a smaller tensile stress state, which would be related to the stress relaxation by the LT-AlN IL.

Figures 2a and b are optical micrographs of samples A and B. As shown in Fig. 2a, the cracks are parallel to [11̅00] in the GaN layer and hence form a network of cracks in three sets of parallel arrays at 120° to one another (sample A). The crack density (the length of crack lines per unit area) was calculated to be 1.87 × 10⁻² µm/µm². However, no cracks were observed on the surface of GaN when the LT-AlN IL...
IL was inserted between GaN layers as shown in Fig. 2b. Cracks in GaN on Si are formed as a result of a large biaxial tensile stress [1], the origin of which is the large difference in the thermal expansion coefficients of GaN and Si(111). Recently, Amano et al. [2] and Han et al. [3] reported that an LT-AlN IL is effective in preventing further accumulation of the tensile stress in subsequent GaN overlayers, thereby suppressing the formation of cracks in GaN and AlGaN layers, where all layers were grown on (0001) sapphire substrates. Taking these results into account, it would be expected that the LT-AlN IL in sample B reduced the tensile stress, resulting in a weak tensile strain state in the GaN overlayer on Si(111) substrates. This explanation is also consistent with the PL results, as described in Fig. 1. A possible mechanism for the relaxation in tensile stress in GaN by the LT-AlN IL will be discussed later, along with the structure of the LT-AlN IL.

Figure 3a shows synchrotron X-ray diffraction profiles around the perfect in-plane GaN (1010) reflection along the $Q_x$ direction (H-scan). The $\theta$-rocking for the AlN (0002) reflection is shown in Fig. 3b. As shown in Fig. 3a, the FWHM of sample B grown with the LT-AlN IL is much less than that of sample A grown without the interlayer. The domain size, $d$, along the in-plane direction of samples A and B was calculated to be 628 Å and 967 Å deduced from the formula $d \approx 2\pi/FWHM$, respectively. This result shows that the defect density near the surface region of sample B is less than that of sample A, which can be attributed to a reduction in the dislocation density by the LT-AlN IL. In addition, the FWHM of the $\theta$-rocking curves across the GaN (0002) reflection decreased from 795.6 arcsec in sample A to 655 arcsec in sample B (data not shown). The FWHM of 655 arcsec is comparable to or even better than that for GaN grown on Si(111) by molecular beam epitaxy (510 ~ 1320 arcsec) [8–10]. These XRD results show that the LT-AlN IL greatly improves the in-plane crystalline quality, as well as the alignment of the GaN domain in the surface normal direction.
It is particularly interesting that the LT-AlN IL, which played an important role in eliminating surface cracks and in a significant improvement in crystallinity and optical properties, consisted of tilted domains (Fig. 3b). As shown in Fig. 3b, two distinct peaks were resolved from the central AlN (0002) peak (i.e., nontilted AlN domains with basal planes parallel to the substrate surface) in sample B and that these are significantly different from the $\theta$-rocking curve of sample A. The two resolved peaks indicate the presence of tilted domains, which have basal planes tilted by the angle of $\pm0.9^\circ$ with respect to the substrate normal direction. Laterally modulated or correlated periodic structures were not shown in the highly defective and disordered LT-AlN IL, confirmed by a cross-sectional TEM analysis. This indicates that a lateral correlation effect is not a possible origin of the two additional peaks in Fig. 3b. Recently, similar crystallographic tilts have been observed in lateral epitaxial overgrowths of GaN [11, 12] and it has been suggested that the possible causes were factors like mask densification, etching and/or chemical instability which could lead to stresses sufficient to induce plastic deformation or a crystallographic tilt at the growth temperatures used, thereby relaxing the stresses in the layer [13]. In the growth of an AlN interlayer at a low temperature of 550 $^\circ$C, a continuous amorphous layer would be expected to be formed; Akasaki et al. [14] reported on the formation of an amorphous-like structure during low-temperature AlN growth (600 $^\circ$C). During a temperature increase to 800 $^\circ$C and a subsequent high-temperature GaN overlayer growth, the amorphous AlN interlayer would be annealed and converted into a crystalline structure, resulting in a relaxation of the LT-AlN IL through the formation of tilted domains induced by a lattice- and/or thermal-mismatched stress. Therefore, a subsequent GaN overlayer would be grown on the relaxed LT-AlN IL under compressive stress at the initial stage of growth, leading to an efficient reduction in tensile stress and the resultant decrease in crack density.

4 Conclusions

The insertion of an LT-AlN IL is reported as an effective way to grow crack-free high-quality GaN layers on Si[111]. Additional distinct domains with a tilt angle of $\pm0.9^\circ$ were found in the LT-AlN IL by synchrotron X-ray diffraction. The LT-AlN IL, relaxed by the formation of tilted domains, appears to play an important role in the relaxation of tensile stress and the reduction of threading dislocations, resulting in significant improvements in the structural and optical properties of GaN.

Acknowledgment

This work was supported by the program on the National Research Laboratory for Nanophotonic Semiconductors in Korea.

References