Journal of the Korean Physical Society, Vol. 50, No. 3, March 2007, pp. 801∼805

Growth of a GaN Epilayer on a Si (111) Substrate by Using an AlN/GaN Superlattice and Application to a GaN Microcavity Structure with Dielectric-Distributed Bragg Reflector

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(Received 12 September 2006)

We have grown GaN films with a thickness of about 1.2 μm on Si (111) substrates by using metalorganic chemical vapor deposition and a AlN/GaN superlattice. In order to optimize the pair number of the AlN/GaN superlattice, we have grown GaN films on Si (111) substrates having various pair numbers of the AlN/GaN superlattices. When the pair number of the AlN/GaN superlattices is 15 pairs, no cracks are observed in the GaN film grown on Si (111), and the GaN film shows the best surface and crystal quality. Also, the GaN micro-cavity structure with SiO_2/ZrO_2 dielectric-distributed Bragg reflectors as both cavity mirrors has been fabricated by means of transferring a InGaN/GaN multiple quantum well structure from the Si (111) substrate onto a sapphire carrier and wet-chemical etching of the Si substrate. Dips in the reflectivity spectrum of the fabricated micro-cavity structure have been observed at wavelengths of 405 nm 433 nm, indicating the resonance mode of the cavity.

PACS numbers: 81.05.Ea, 81.15.Gh, 68.55.-a, 85.60.Bt, 42.55.Sa
Keywords: GaN, Superlattice, MOCVD, Microcavity

I. INTRODUCTION

In recent years, the III-nitrides and their alloys with unique properties, such as large bandgap energy, high thermal conductivity, and considerable hardness, have attracted much attention as promising materials for use in optical display, data storage, and high-power and high-frequency electronic devices [1–6]. Commercially available nitride devices, such as light-emitting diodes (LEDs), are usually grown on sapphire or SiC substrates. Another attractive candidate for the substrate material for GaN growth is Si. Si is a very promising substrate material for the growth of GaN layer, allowing future integration of well-established Si electronics with GaN-based photonic devices. Si as a substrate for GaN growth has many advantages, such as high quality, large size, low cost, and thermal stability at high growth temperature [7]. Several groups have succeeded in fabricating GaN-based photonic and electronic devices, such as LEDs, ultraviolet detectors, and high electronic mobility heterostructures, on Si substrates [8–13]. However, the large lattice (∼17 %) and thermal mismatches (56 %) between GaN and Si yield a biaxial tensile stress in the GaN/Si interface. This residual tensile stress leads to misfit dislocations and cracks in the GaN epilayer. Recently, several groups have attempted to grow GaN films by introducing various intermediate layers and growth techniques, such as AlN, GaN, and Al(Ga)N/GaN superlattices (SLs), laterally epitaxial overgrowth, and selective area growth [14–24].

In this work, we report on the growth of a crack-free GaN epilayer on a Si (111) substrate by using AlN/GaN SLs and metalorganic chemical vapor deposition (MOCVD). Also, the effect of the pair number of SLs on the GaN epilayer is discussed based on the results of Raman spectroscopy, optical microscopy, atomic force microscopy (AFM), and high-resolution X-ray diffraction (HRXRD) results. Also, for application to resonant-cavity LED and vertical-cavity surface-emitting lasers (VCSELs), we fabricated a GaN-based microcavity structure with SiO_2/ZrO_2 dielectric-distributed bragg reflectors (DBRs).

II. EXPERIMENT

GaN epilayers were grown on Si (111) substrates by using MOCVD. Trimethylgallium (TMGa), trimethylaluminum (TMAl), and ammonia (NH_3) were used as the source precursors for Ga, Al, and N, respectively. The substrates were further *in-situ* annealed for two minutes

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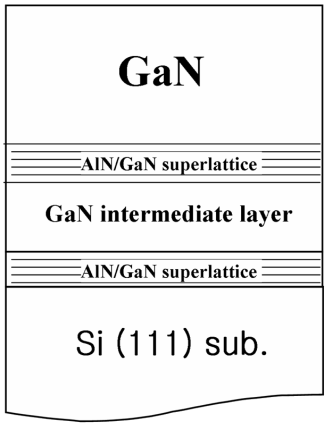

Fig. 1. Schematic cross section of the GaN epilayer grown on a Si (111) substrate.

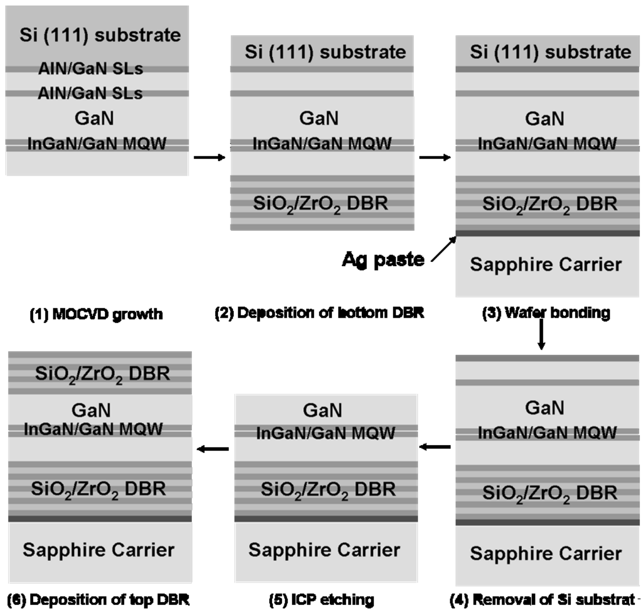

Fig. 2. Process flow chart of the GaN microcavity structure.

at 1070 °C in a H_2 ambient followed by exposure to a TMAl flow rate of 12 μmol/min for 10 seconds in order to prevent the formation of a Si_xN_y layer [25, 26]. The pressure and the temperature were kept at 100 Torr and 1070 °C during all growth steps, respectively. As Fig. 1 shown, prior to the deposition of the GaN epilayer, a strain compensation structure consisting of a AlN/GaN(2/5 nm) SL, a GaN intermediate layer, and a AlN/GaN(2/5 nm) SL was grown on a Si (111) substrate to efficiently reduce the tensile stress between the Si substrate and the GaN epilayer. The TMGa, TMAl, and NH_3 flow rates were 90 μmol/min, 12 μmol/min, and 2800 sccm, respectively. Finally, a GaN epilayer ($\sim$1.2 μm) was grown by feeding TMGa and NH_3 with flow rates of 90 μmol/min and 2800 sccm, respectively. To optimize the pair number of the AlN/GaN SL, we varied

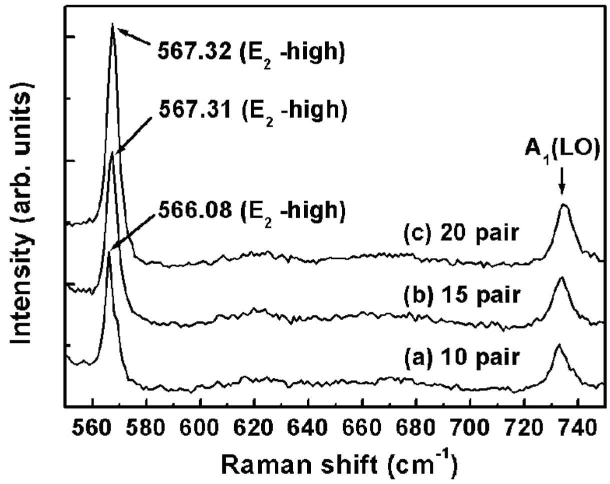

Fig. 3. Raman spectra for GaN epilayers grown on Si (111) substrates with (a) 10-pair, (b) 15-pair, and (c) 20-pair AlN/GaN SLs.

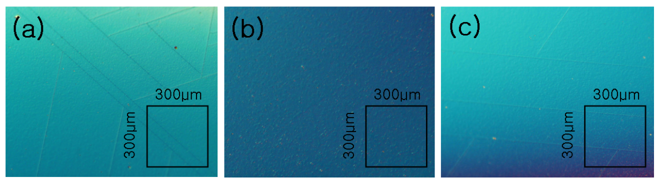

Fig. 4. Optical microscope images of the surfaces of GaN epilayer on Si (111) substrates with (a) 10-pair, (b) 15-pair, and (c) 20-pair AlN/GaN SLs.

the pair number of the SL from 10 to 20 while keeping the other growth parameters constant. The thickness of the GaN intermediate layer was fixed to be 300 nm.

For the GaN microcavity structure, five periods of the InGaN/GaN MQW structure as an active layer was grown on a crack-free GaN eplayer grown on Si (111) at optimized condition, and the undoped GaN cap layer was grown. The MQW structure was transferred from the Si substrate onto the sapphire carrier as shown in Fig. 2. First, after the deposition of 15 pairs of SiO_2/ZrO_2 DBRs on the GaN cap layer, the backside of the Si substrate was mechanically polished to a thickness of about 100 μm. The InGaN/GaN MQW structure, with 15 pairs of SiO_2/ZrO_2 DBRs, was then bonded onto the surface of the sapphire carrier by using silver paste as the bonding medium. By wet-chemical etching in a HNA solution (HF : HNO_3 : CH_3OOH = 1 : 1 : 1), the Si substrate was removed completely. Next, the AlN/GaN SL-GaN-AlN/GaN SL buffer structure was etched using an inductive coupled plasma (ICP) because of its low crystal quality. In order to improve the surface roughness of the exposed region, we attempted a surface treatment by using an ICP etcher. Finally, 10 pairs of SiO_2/ZrO_2 DBRs were deposited as a top mirror by using an electron-beam evaporator. The target center wavelength was 415 nm.

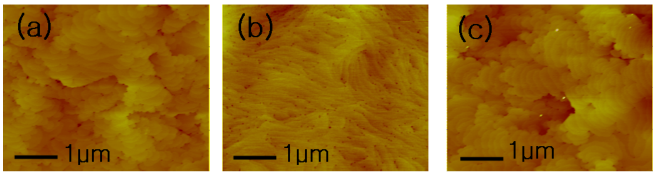

Fig. 5. AFM surface images of GaN epilayers grown on Si (111) substrates with (a) 10-pair, (b) 15-pair, and (c) 20-pair AlN/GaN SLs.

III. RESULTS AND DISCUSSION

In order to investigate the influence of the pair number of the AlN/GaN SL, we varied the pair numbers of the lower and the upper SLs from 10 to 20 pairs while keeping the thickness of the GaN intermediate layer at 300 nm. To understand and minimize the stress due to mismatches in the lattice constants and thermal expansion coefficients, we calculated the amount of residual stress in the samples by using the measured frequency shifts of the E_2-high mode in the Raman spectra [27]. Fig. 3 shows the room-temperature Raman spectra of the E_2 (TO)-high line and A1 (LO) line from the GaN epilayers grown on Si (111) substrates with (a) 10-pair, (b) 15-pair, and (c) 20-pair AlN/GaN SLs. With increasing pair number of the AlN/GaN SL from 10 to 15, both the TO and the LO phonon peaks shifted to higher frequencies from 566.08 cm^{-1} and 732.89 cm^{-1} to 567.31 cm^{-1} and 734.12 cm^{-1}, respectively. The values of the tensile stress, which were calculated using the E_2 phonon peak observed at 568 cm^{-1} for the stress-free GaN and the relation $\Delta\omega = k\sigma_{xx}$ cm^{-1} GPa^{-1} [28,29], are 0.45 GPa and 0.16 GPa for 10-pair and 15-pair SLs, respectively. Here, $\Delta\omega$ is the phonon peak shift, σ_{xx} is the biaxial stress, and k (=4.3) is the pressure coefficient [30]. The GaN epilayer with 15-pair AlN/GaN SLs shows a significantly reduced in-plane stress as compared to the GaN epilayer with 10-pair AlN/GaN SLs. Meanwhile, 15-pair and or 20-pair SLs produce almost the same stress in the final GaN epilayer.

Fig. 4 shows optical microscope images of the surfaces of GaN epilayers grown on Si (111) substrates with (a) 10-pair, (b) 15-pair, and (c) 20-pair AlN/GaN SLs. With increasing pair number of the AlN/GaN SL from 10 to 15, no cracks are observed in the GaN epilayer. However, cracks are observed when the pair number of the AlN/GaN SL increases to 20. This result is consistent with the AFM result. Fig. 5 shows the AFM surface images of the GaN epilayers grown on Si (111) substrates with (a) 10-pair, (b) 15-pair, and (c) 20-pair AlN/GaN SLs. The rms values of the surface roughness are (a) 0.49 nm, (b) 0.26 nm, and (c) 0.61 nm, respectively. These values are comparable to those obtained for GaN on sapphire. When changing the pair number of the AlN/GaN SL from 10 to 15, the rms value decreases whereas AlN/GaN SLs of 20 pairs causes the GaN epilayer to become rougher.

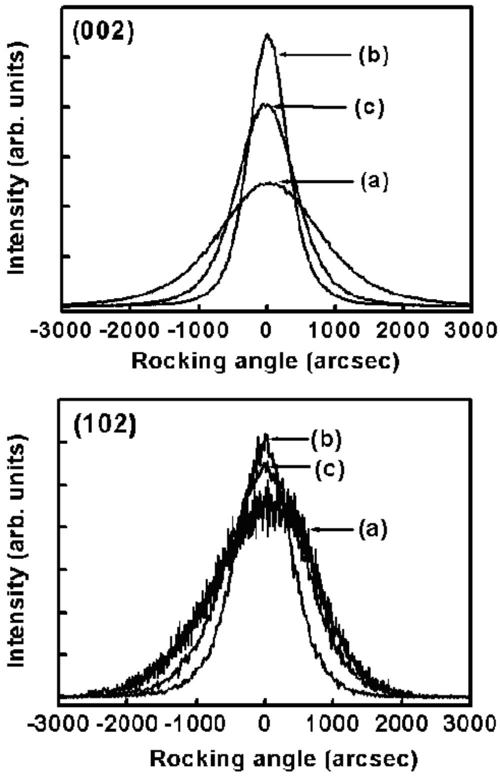

Fig. 6. HRXRD rocking curves of GaN epilayers grown on Si (111) substrates with (a) 10-pair, (b) 15-pair, and (c) 20-pair AlN/GaN SLs: (002) symmetry plane (top), (102) asymmetry plane (bottom).

Fig. 6 shows HRXRD rocking curves (ω-scans) of (002) symmetry planes and (102) asymmetry planes of GaN epilayers grown on Si (111) substrates with (a) 10-pair, (b) 15-pair, and (c) 20-pair AlN/GaN SL. As the figure shown, the full widths at half maximum (FWHM) of HRXRD rocking curves for the (002) and the (102) planes of GaN epilayers grown with AlN/GaN SLs for 10 pairs, 15 pairs, and 20 pairs are (a) 1350 and 1466, (b) 608 and 824, and (c) 885 and 1170 arcsec, respectively, which means that the GaN epilayer with the 15-pair AlN/GaN SL has the highest crystal quality. This behavior of crystal quality is very similar to the change in the surface property shown in Fig. 4 and Fig. 5. From these results for the surface and the crystal qualities, we found that the 15-pair AlN/GaN SL gave rise to a significant improvement in the quality of the GaN epilayer grown on a Si(111) substrate.

Now, we set the pair number of the AlN/GaN SL to 15 and grew a five-period InGaN/GaN MQW structure on a GaN/Si (111) substrate. A GaN microcavity structure with SiO_2/ZrO_2 dielectric-distributed Bragg reflectors was fabricated by means of transferring the InGaN/GaN MQW structure from the (111) Si substrate onto a sapphire carrier and wet-chemical etching of the substrate. Fig. 7 shows a cross-sectional scanning electron microscope (SEM) image of the fabricated GaN micro-cavity

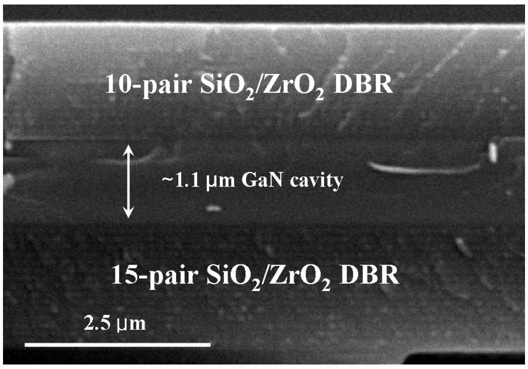

Fig. 7. Cross-sectional scanning electron microscope (SEM) image of the fabricated GaN micro-cavity structure.

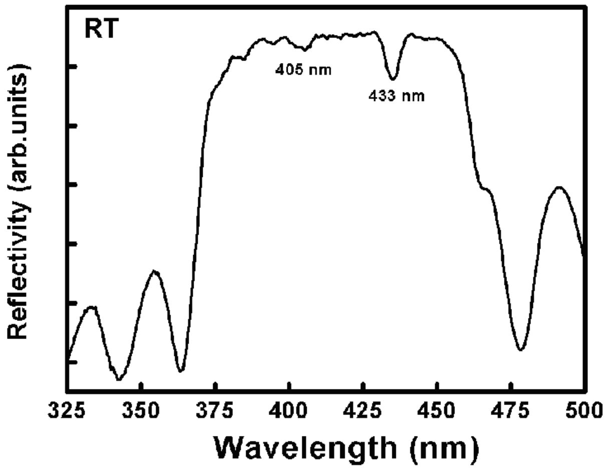

Fig. 8. Measured reflectivity spectrum of the fabricated microcavity structure.

structure with 15 (bottom)/10 (top) pairs of dielectric DBRs. The SiO_2/ZrO_2 interfaces seem to be well defined. The peak reflectivities were about 98 % and 99 % for the top and the bottom dielectric DBRs, respectively.

Fig. 8 shows the reflectivity spectrum of the complete GaN micro-cavity structure at room temperature. The center of the stop-band of the reflectivity spectrum is about 415 nm. The stop-band width of about 90 nm is larger than that of the GaN-based reflectors due to the relatively high refractive index contrast between dielectric materials. Cavity resonances are clearly observed at wavelengths of 405 nm and 433 nm for this micro-cavity structure. However, due to the disagreement between the calculated and the experimental cavity lengths, the position of the cavity resonance mode is not matched to the center of the stop-band of the dielectric DBRs.

IV. CONCLUSIONS

We have successfully grown a crack-free GaN epilayer on a Si (111) substrate by using MOCVD and AlN/GaN SLs, and we fabricated a GaN microcavity structure with dielectric DBRs. The GaN microcavity structure with SiO_2/ZrO_2 dielectric-distributed Bragg reflectors was fabricated by means of transferring a InGaN/GaN multiple quantum-well structure from a (111) Si substrate onto a sapphire carrier and wet-chemical etching of the substrate. Cavity resonances are clearly observed at wavelength of 405 nm and 433 nm. The result provides evidence for the usefulness of this method for realizing a GaN based VCSEL on a Si (111) substrate.

ACKNOWLEDGMENTS

This work was supported by a Korea Research Foundation grant (R05-2004-000-10396-0).

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