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Communication

A Novel Technique for Growing Crack-Free GaN Thick Film by Hydride Vapor Phase Epitaxy

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To prevent the cracking of GaN thick films grown on a sapphire substrate by hydride vapor phase epitaxy (HVPE), a novel technique without complex processes is developed. By adding a temperature ramping step in the HVPE GaN epitaxy process, more than 300-μm-thick high-quality crack-free GaN thick films on sapphire substrate can be obtained by this technique. After separation by a conventional laser-induced lift-off process, a 1.5 in. 300 μm freestanding GaN wafer with a dislocation density of approximately $1 \times 10^7\,cm^{-2}$ could be fabricated without any cracks. No additional designed-patterned or stress-reduced structures were applied in these samples to reduce the dislocation density and thermal stress.
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KEYWORDS: GaN, HVPE, freestanding GaN, CL, TEC

GaN substrates are highly promising for applications in UV light-emitting diodes, laser diodes, and high-power and high-current density nitride-based devices.[1–5] Given the difficulty in obtaining high-quality GaN substrates, most current nitride-based devices are heteroepitaxially grown on lattice-mismatched substrates, such as Al_2O_3 (sapphire), Si, GaAs, and SiC. The differences in thermal expansion coefficient (TEC) and lattice mismatch between GaN and foreign substrates usually induce a large residual strain in GaN thick films. Such a strain normally results in GaN films cracking in the epitaxy or cooling process from growth temperature to room temperature.[6–10]

The low decomposition temperature compared with the melting temperature of GaN makes it very challenging to obtain GaN by liquid-phase growth technology.[11] Hydride vapor phase epitaxy (HVPE) is the most effective method of fabricating a GaN substrate owing to its fast GaN growth rate.[12–15] In most cases of HVPE GaN growth, the foreign substrate used is a sapphire substrate owing to its reliability and low cost. The lattice mismatch and the difference in TEC between sapphire and GaN are 16% and roughly 2×10^{-6}/K, respectively.[16–18] If the growth temperature of HVPE GaN is near 1000 °C, the difference in TEC between sapphire and GaN is 0.2%, which results in the GaN/sapphire structure bending and cracking during the cooling process.

Several groups have developed self-separation techniques for producing freestanding GaN thick films by HVPE via several methods such as void-assisted separation, facet-controlled separation, and $WSiN_x$ epitaxial lateral overgrowth techniques.[19–22] These self-separation techniques can prevent GaN thick films from cracking; however, they require excess processing, which increases the cost of GaN thick-film epitaxy. In addition, the yield of self-separation techniques for obtaining complete freestanding GaN wafers without any cracks is still very low.

In this investigation, a very simple technique without complex processes was developed to prevent cracks caused by differences in TEC between GaN and original substrates for HVPE growth.

A 4-μm-thick GaN layer was initially grown on a *c*-plane sapphire substrate as template for the HVPE GaN thick-film regrowth by metal–organic chemical vapor deposition (MOCVD). The GaN template was mounted a horizontal HVPE reactor to grow a 300-μm-thick GaN film. The HVPE GaN growth temperature was started at a low temperature (LT) of 950 °C and ramped to a high temperature (HT) of 1050 °C at a ramping rate of about 1 °C/min. After the temperature ramping, GaN growth temperature was maintained at 1050 °C until the thickness of the GaN film exceeded 300 μm. In the GaN growth process, NH_3 gas was used as the nitrogen source, and GaCl, generated by liquid gallium and HCl gas at 850 °C, was used as the gallium source. The ambient of the carrier gas was a mixture of H_2 and N_2, and pressure was maintained at 900 mbar throughout the GaN growth process. To provide a significant comparison, another GaN sample was directly grown on the MOCVD template at 1050 °C without the temperature ramping step and LT layer.

Figure 1 shows the full structure of the GaN film grown with the temperature ramping step. The HVPE GaN growth process was started at 950 °C for rough 50 μm GaN growth. Then, the growth temperature was ramped at 1 °C/min until it reached 1050 °C. The thicknesses of the GaN film grown with temperature ramping and at HT were roughly 120 μm and over 130 μm, respectively.

As can be seen in Fig. 2(a), the sample grown with the temperature ramping step shows crack-free and mirrorlike features on the surface. However, the sample grown without the temperature ramping step and LT layer was cracked, as shown in the inset of Fig. 2(a). The 1.5 in. crack-free sample was separated by a laser lift-off process using a 355 nm Nd:YAG laser, as shown in Fig. 2(b). The HVPE GaN was characterized by scanning electron microscopy (SEM), cathodoluminescence (CL) measurement, high-resolution X-ray diffraction (HRXRD) analysis, micro-Raman spectroscopy, and etch pit density (EPD) measurement. The sample for the EPD experiment was etched at 220 °C for 20 min in a mixture of H_3PO_4 and H_2SO_4 at a ratio of 1 : 3. To understand the stress distribution, the CL and Raman measurements were carried out on the GaN thick films samples without separation from the sapphire substrate. The

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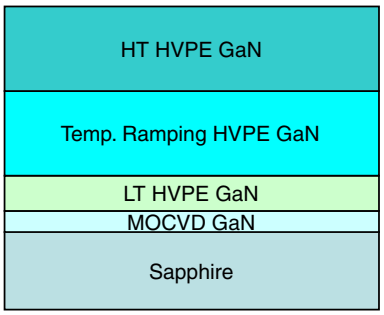
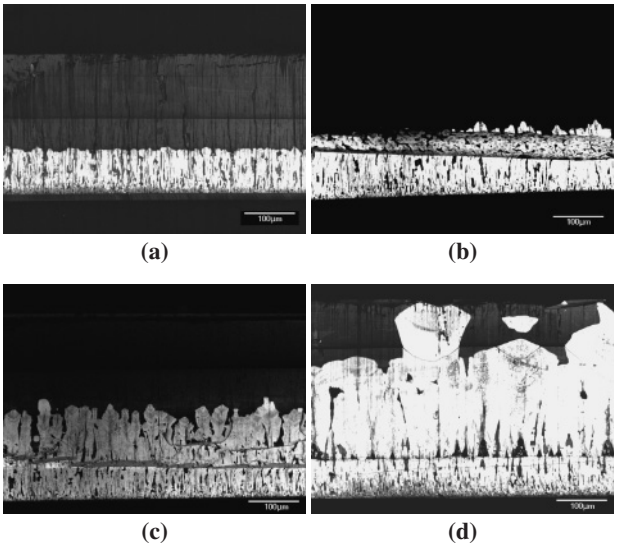


Fig. 1. (Color online) Schematic of the temperature ramping GaN structure grown by HVPE.

(a) (b)

Fig. 2. (Color online) (a) A 1.5 in. 300 μm crack-free GaN thick film grown on sapphire substrate with temperature ramping step. The inset is the cracked GaN grown without any temperature ramping step. (b) A 300 μm crack-free freestanding GaN separated by laser lift-off process.



(a) (b) (c) (d)

Fig. 3. Cross-sectional CL images of GaN films grown with temperature ramping layer at various ramping rates: (a) 3.3, (b) 1.7, (c) 1.0, and (d) 0.8 °C/min.

XRD analysis was carried out before and after the laser lift-off process.

Figure 3 shows the cross-sectional CL contrasted images of the HVPE GaN samples grown at different ramping rates. A bright band was clearly observed in these samples with different thicknesses. Several reports have mentioned that the bright band corresponds to free carriers, which result from defects and impurities.[21,23] Figure 3(a) shows the sample grown with a ramping rate of 3.3 °C/min, and the thickness of the bright region is 90 μm. Figures 3(b)–3(d) show the samples with ramping rates of 1.7, 1, and 0.8

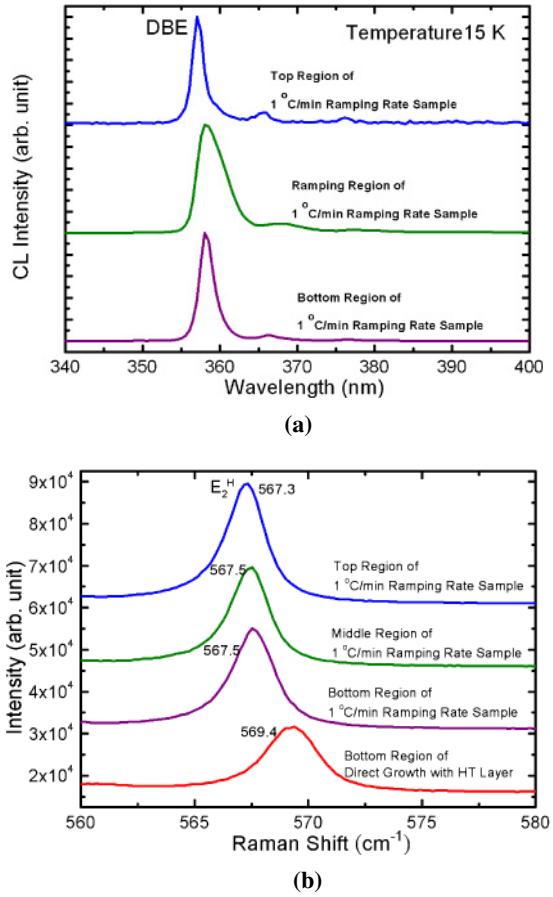


Fig. 4. (Color online) (a) CL spectra of cross-sectional regions of the sample grown with 1.0 °C/min ramping rate. The spectra were measured in the bottom, ramping, and HT regions. (b) Raman spectra of cross-sectional regions of the same sample in (a). The spectra were measured in the bottom, ramping, and HT regions.

°C/min, respectively. As the ramping rate decreases, the ramping process needs more time to achieve HT growth. This means that thicker HVPE GaN was grown during the lower-temperature process than during the HT, high-quality growth process, as ramping time was increased. The of LT growth and ramping growth regions were like a large buffer layer that relaxed the thermal stress to prevent the cracking of GaN thick films grown on sapphire substrate. In this paper, the discussion mainly focuses on the sample of 1 °C/min ramping rate; results obtained with different ramping rates will be compared and discussed in the future.

Figure 4(a) shows the low-temperature CL spectra of a cross-sectional region of the sample grown with a 1 °C/min ramping rate at 15 K. The CL spectrum data were measured at the bottom, middle, and top regions of the cross-sectional sample. The middle and top regions corresponded to the ramping layer and HT growth layer, respectively. Near-band-edge emission peaks were observed at 358 nm (3.46 eV), 358 nm, and 357 nm (3.47 eV), which correspond to the bottom, ramping, and HT regions, respectively, for HVPE GaN growth. A biaxial stress of one GPa would shift the near-band-edge peak by 27 ± 2 meV. Tensile or compressive stress can be calculated with reference to the strain-free emission peak at 3.467 eV.[24] If the near-band-edge emission energy was less than 3.467 eV, the presence of tensile stress would be expected; on the other hand, larger values would

indicate that the stress was compressive. On the basis of the report by Kisielowski *et al.*,[24] the stress in the bottom, ramping, and HT regions of the GaN/sapphire structure could be calculated. The bottom and ramping regions showed a small tensile stress of 0.1 GPa, and the HT region showed a compressive stress of 0.2 GPa. Normally, compressive stress would be observed in GaN/sapphire structures; the presence of tensile stress at the bottom and ramping regions may be in the error range of the CL measurement.

Raman scattering spectra were also measured at the same positions as CL spectra at room temperature, as shown in Fig. 4(b). The peaks of the E_2 (high) mode phonon frequency were observed at 567.5, 567.5, and 567.3 cm^{-1} in the bottom, ramping, and HT regions, which corresponded to compressive stresses of 0.12, 0.12, and 0.07 GPa, respectively.[25] The E_2 (high) mode phonon frequency of the sample grown without ramping and the LT layer was observed at 569.2 cm^{-1} in the bottom region of GaN, which indicated a larger compressive stress of 0.5 GPa.

From the results of the CL and Raman spectrum analyses, we determined that residual stress could be relaxed in the structure of GaN thick films without separation from the sapphire substrate by the temperature ramping technique. The relaxation of thermal stress is useful for preventing the cracking of the GaN/sapphire structure. This is very important for successfully fabricating GaN substrates by HVPE.

Figure 5(a) shows the full width at half maximum (FWHM) of the HRXRD rocking curve of the GaN symmetric (002) plane before and after the laser lift-off process. The FWHM of GaN on the sapphire substrate is 847.2 arcsec. This large value is due to the bowing of the GaN/sapphire structure. The FWHM of the freestanding GaN thick film is 156.7 arcsec, which could be more improved after the LT growth region was removed. Figure 5(b) shows a top-view CL image of the 300 μm free-standing GaN. The dislocation density is about 1×10^7 cm^{-2} by the calculation of both the CL image and the EPD measurement. The random distribution of dark spots in the CL image shows that there is no additional designed-patterned structure applied to reduce dislocation density in these samples.

In summary, a very simple technique without complex processes for preventing cracks in GaN thick films grown by HVPE is developed. High-quality 1.5 in. large-area free-standing GaN films with thicknesses of over 300 μm can be obtained by this technique after the consequent lift-off process. The FWHM of HRXRD of GaN(002) is 156.7 arcsec and the dislocation density is about 1×10^7 cm^{-2}. No additional designed-patterned structure was required in these samples to reduce dislocation density and thermal stress.

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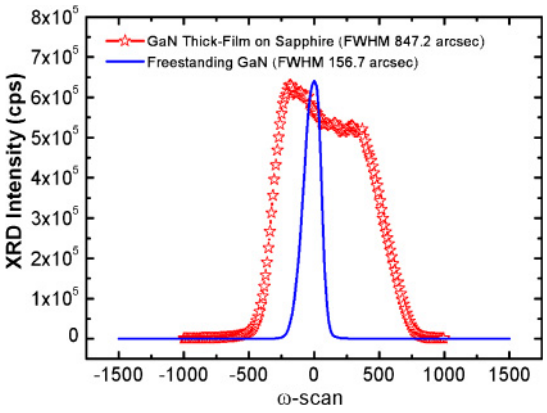
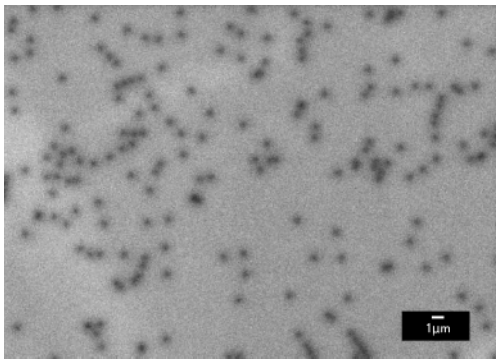


(a)



(b)

Fig. 5. (Color online) (a) Rocking curve of HRXRD of GaN grown with a 1.0 °C/min ramping rate before and after laser lift-off process. The FWHM is 156.7 (847.2) arcsec after (before) the laser lift-off process. (b) Top view CL image of freestanding GaN wafer. The dark spots show the positions of dislocations.

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