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Strain-reduced GaN thick-film grown by hydride vapor phase epitaxy utilizing dot air-bridged structure

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ABSTRACT

A 300 μm GaN thick-film, in diameter 1.5 in, was demonstrated without any crack by hydride vapor phase epitaxy (HVPE) growth. The technique used in relaxing the residual stress caused by differences of thermal expansion coefficients (TEC) and lattice constants between GaN and sapphire substrate to prevent GaN film from crack is called a dot air-bridged structure. After the laser lift-off process, 300-μm-thick freestanding GaN wafer, in diameter 1.5 in, could be fabricated. The compressive stress in the dot air-bridged structure was measured by micro-Raman spectroscopy with the E_2(high) phonon mode. The compressive stress could be reduced to as small as 0.04 GPa, which could prevent the crack during the epitaxial process for GaN growth by HVPE. It is important to obtain a large-area crack-free GaN thick-film, which can be used for fabricating freestanding GaN wafer.

1. Introduction

The GaN substrate is a highly promising substrate for applications in blue laser diodes, UV light-emitting diodes, and high-power and high current density optoelectronic devices, which are sensitive in dislocation density or thermal conduction [1–4]. However, due to the difficulty of obtaining high-quality and reasonable cost GaN substrates, most current nitride-based optoelectronic devices are heteroepitaxially grown on lattice-mismatched substrates, such as sapphire (Al_2O_3), 6H-SiC, GaAs, and Si substrates. The large mismatches in lattice constants between the foreign substrates and epitaxial GaN films generate high-density dislocations. Furthermore, the difference between the thermal expansion coefficients (TEC) of foreign substrate and epitaxial GaN films causes large bowing of the epi-layers and the substrates. The bowing usually induces large residual strain in epitaxial GaN films, especially, in a GaN thick-film grown by hydride vapor phase epitaxy (HVPE). The large residual strain normally makes GaN thick-film crack in epitaxy or in the cooling down process from the growth temperature to room temperature. When crack occurred it is impossible to get the complete GaN wafer from the foreign substrate, e.g. with laser lift-off or self-separation techniques. However, only few methods can be used in reducing thermal stress for the GaN epitaxial procedure. Dam et al. [5] reported a gallium-treatment step technique to grow a crack-free GaN layer on sapphire substrates with varying buffer thicknesses of metal-organic chemical vapor deposition (MOCVD) GaN template.

In this investigation, a specially designed epitaxial lateral overgrowth (ELOG) process, called dot air-bridged growth, has been developed to grow thick GaN layers by the HVPE system, which could reduce the residual thermal stress of GaN and foreign substrates. The stress was estimated according to the E_2(high) phonon frequency using micro-Raman scattering spectroscopy.

2. Experiments

Fig. 1 illustrates the HVPE GaN fabrication process of a dot air-bridged structure. First, a 3-μm-thick GaN template was grown by MOCVD on a 330-μm-thick *c*-plane sapphire substrate, 2 in in diameter, for HVPE regrowth. A thin SiN_x layer was deposited on the MOCVD GaN template by plasma-enhanced chemical vapor deposition (PECVD). Then, the conventional photolithographic technique was applied to fabricate a hexagonally aligned dot pattern of 3-μm-wide square seed regions and 4-μm-wide grooves between adjacent square regions. These dot patterns worked as hard mask for subsequent inductively coupled plasma

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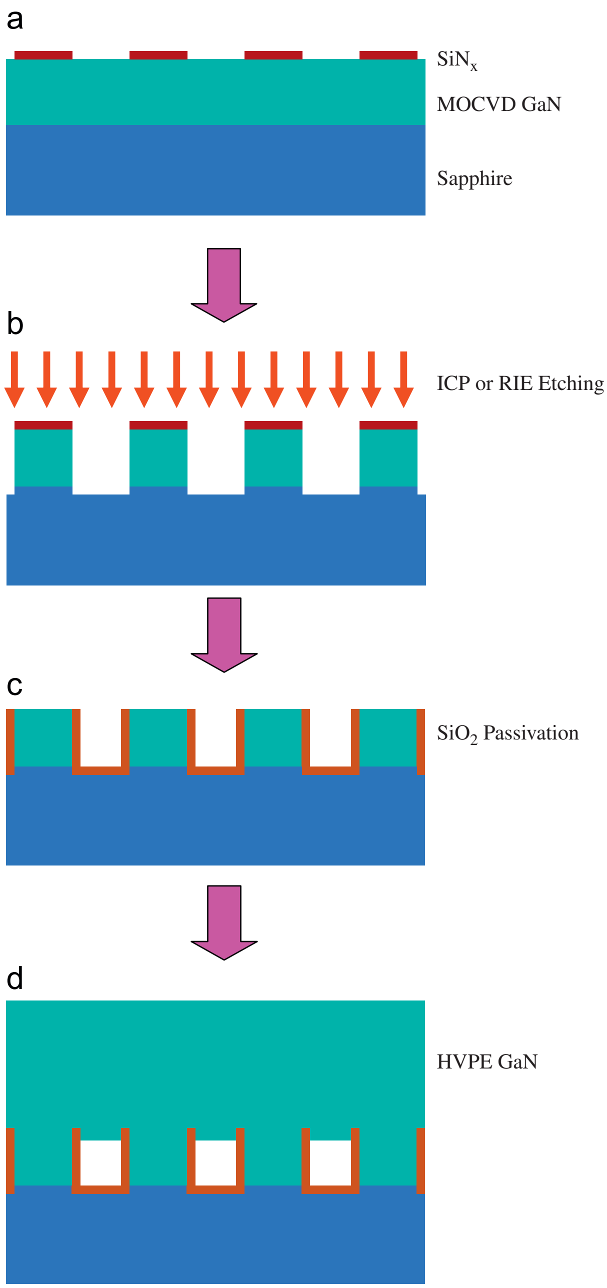

Fig. 1. Schematic of the GaN thick-film fabrication process with the dot air-bridged structure by HVPE. (a) MOCVD GaN template with SiN_x hard mask, (b) dry etching process by ICP, (c) the sidewall passivation of GaN seed regions and (d) GaN thick-film regrowth by HVPE.

(ICP) dry etching. The 3-μm-wide square GaN seed and 4-μm-wide grooves with 200 nm depth etched into the sapphire substrate were formed after ICP dry etching as shown in Fig. 1(b). After the dot air-bridged pattern was defined, a 200 nm dielectric layer was deposited on the sidewall of the GaN seed region by PECVD as shown in Fig. 1(c). Then, the sample was mounted in an HVPE reactor to grow GaN thick-films as shown in Fig. 1(d). For HVPE GaN thick-film growth, the NH_3 gas was used as a nitrogen source and GaCl generated by liquid gallium with HCl gas at 850 °C was used as a gallium source. The ambience of the carrier gas was a mixture of H_2 and N_2. Growth was performed by an Aixtron 800064 horizontal reactor at approximately 1050 °C under 0.9 Pa at a growth rate of about 150 μm/h. The HVPE GaN was characterized by scanning electron microscopy (SEM), cathodoluminescence (CL), and micro-Raman spectroscopy.

3. Results and discussion

Fig. 2(a) shows the bird's-eye view SEM image of a GaN template of a dot air-bridged structure. The region of GaN seeds was isolated clearly and arrayed in hexagonal geometry. Fig. 2(b) shows the bird's-eye view SEM image of initial growth for 2 min by HVPE. The morphology of the initial state of the dot air-bridged structure in the GaN seed region formed a pyramid shape. The shape and the size distribution of the pyramid-shaped GaN islands were uniform. The pyramid-shaped GaN islands were formed by the $\{10\bar{1}1\}$ crystal facets, which were stable, and the growth rate of this facet was very slow due to the low surface energy [6,7]. The stability of these crystal facets makes the GaN epitaxial layer harder to coalesce along the $\langle 10\bar{1}0\rangle$ direction than along the $\langle 11\bar{2}0\rangle$ direction, which was the normal coalesced direction for ELOG. If the distance of the groove region is too large, the GaN film is very hard to coalesce to form a complete thick-film. Then, polycrystalline GaN will grow in the large groove region. The dot air-bridged structure was not as good as the stripe air-bridged structure in a large groove distance in dislocation reduction. However, a large decrease was still observed in the dislocation density of a dot air-bridged structure compared with no-structure samples. Fig. 3(a) shows the plane-view CL image of the dot air-bridged structure after the HVPE thick-film regrowth. The dark spots indicated the position of dislocations with the density of $1.8\times10^{7}\,cm^{-2}$, which is much smaller than the direct growth of GaN on sapphire substrates without any structure of patterns. The full-width-at-half-maximum (FWHM) of CL spectra at 300 K of the dot air-bridged sample was also measured. The FWHM measured from the surface of the dot air-bridged sample was 77 meV without an observed yellow band. Fig. 3(b) shows the cross-sectional SEM image of the pyramid-shaped GaN islands. The dashed line of GaN islands indicated a special growth region in Fig. 3(b). In these regions, the GaN was grown toward the $\langle 000\bar{1}\rangle$ direction and the high-index crystal facets were observed. This phenomenon resulted from the slow growth rate of the $\{10\bar{1}1\}$ crystal facets of pyramid-shaped GaN islands, which make the GaN have enough time to grow towards the $\langle 000\bar{1}\rangle$ direction in the dash-line regions before the pyramid-shaped GaN islands coalesced together. The GaN grown towards the $\langle 000\bar{1}\rangle$ direction changed the height in the void of the dot air-bridged structure. The height of the void in the dot air-bridged structure could determine whether the HVPE GaN thick-film can self-separate from the sapphire substrate or not. If the HVPE GaN growth conditions and the process of fabricating a dot air-bridged structure were controlled well, the GaN growth towards the $\langle 000\bar{1}\rangle$ direction would be much decreased, and the GaN thick-film would be easily separated from the sapphire substrate. Otherwise, the HVPE GaN thick-film would remain on the sapphire substrate without separation. Even if the GaN thick-film was not separated from the sapphire, the residual stress was still relaxed without cracking the GaN. Crack-free freestanding GaN wafer can still be fabricated by the laser lift-off technique. In this paper, the discussion mainly focuses on the samples without any separation from the sapphire substrate, and the self-separation technique will be discussed in the other paper.

To understand the residual stress distribution, a 100-μm-thick GaN sample with the dot air-bridged structure on a sapphire substrate was cleaved to measure the cross-sectional stress variation. Fig. 4(a) shows the cross-sectional CL image of the dot air-bridged structure. The height of voids in this sample between the interface of the GaN and the sapphire was roughly 0.4 μm,

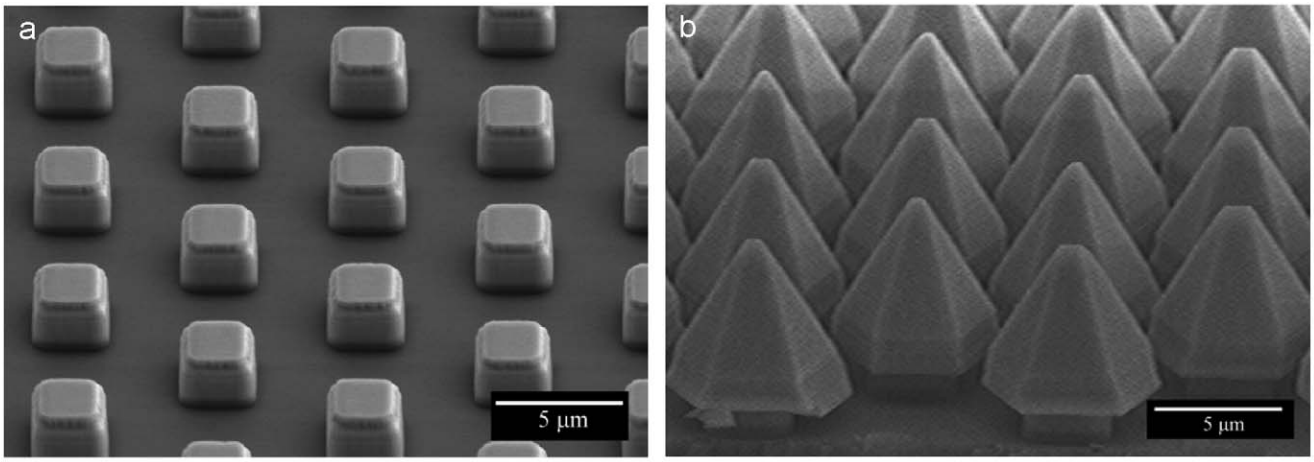

Fig. 2. (a) The bird's-eye view SEM image of the dot air-bridged structure and (b) the SEM image of 2 min initial growth state of the dot air-bridged structure by HVPE.

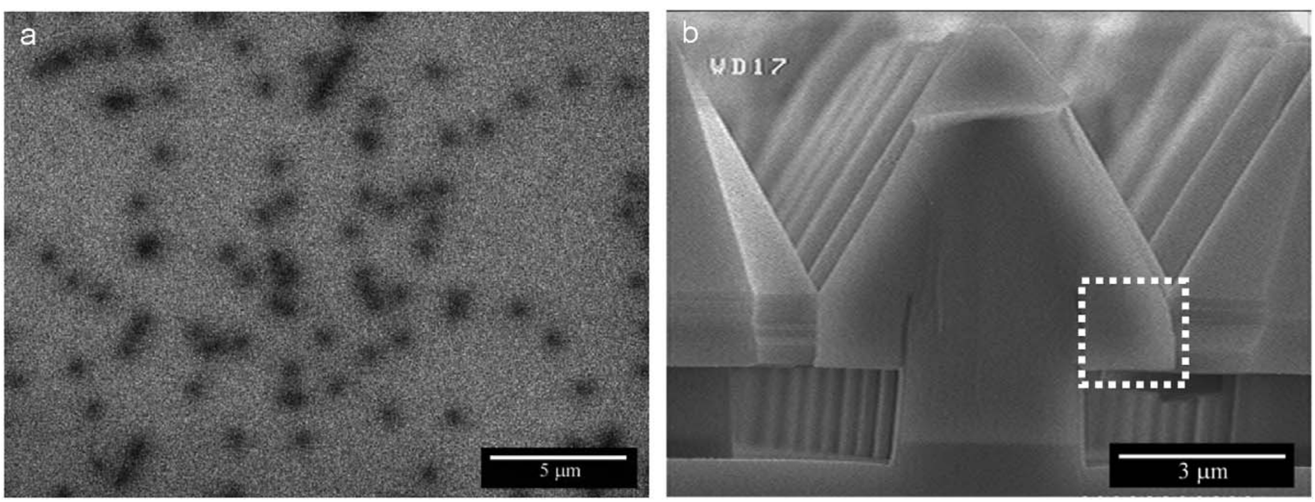

Fig. 3. (a) The plane-view CL image of GaN thick-film of the dot air-bridged structure by HVPE and (b) the cross-sectional SEM image of GaN islands of the dot air-bridged structure. The dash-line region indicates the GaN grown towards the $\langle 000\bar{1}\rangle$ direction.

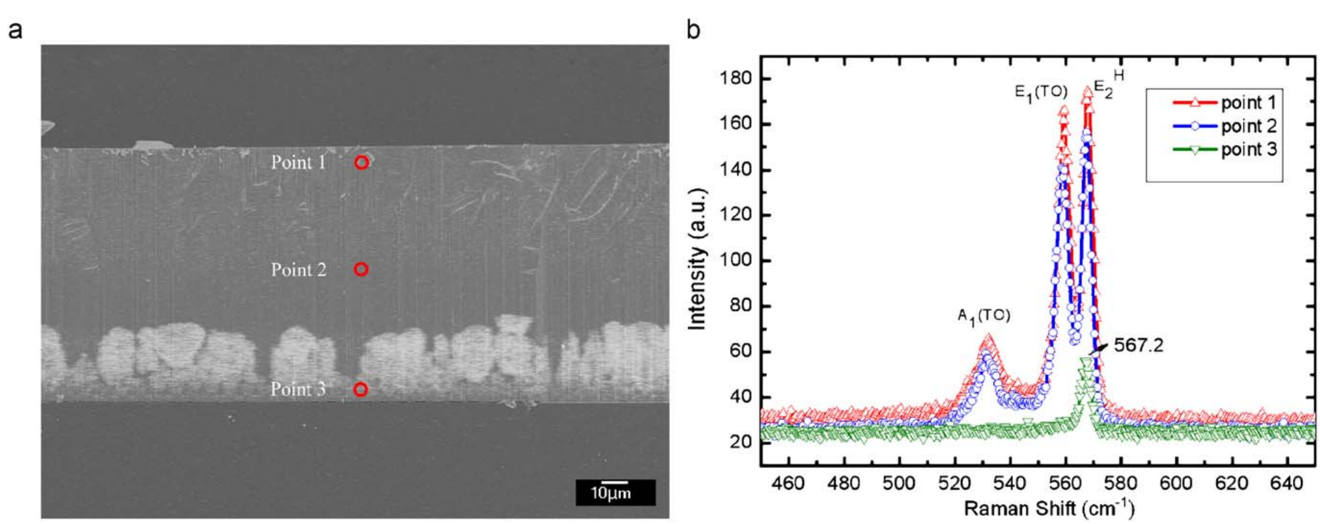

Fig. 4. (a) Cross-sectional CL image of the dot air-bridged sample and (b) Raman spectra recorded on points 1, 2, and 3 of (a).

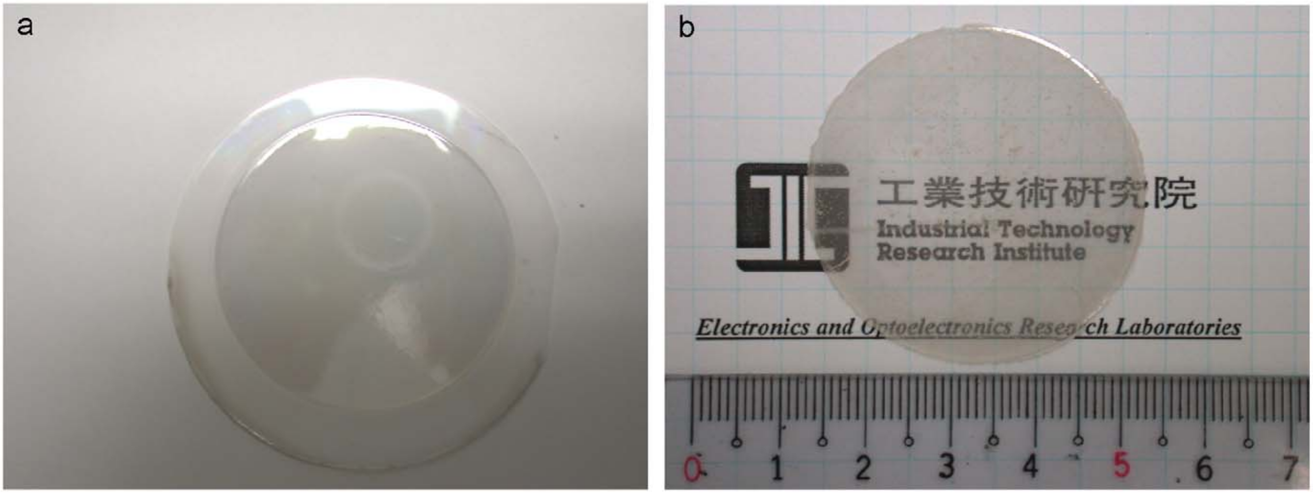

Fig. 5. (a) A 1.5-in-diameter, 300 μm GaN thick-film on a 2 in sapphire substrate without any crack and (b) the 1.5 in-diameter 300 μm freestanding GaN wafer without any crack. The GaN wafer is transparent.

which was due to the $\langle 000\bar{1} \rangle$ growth of the GaN. The bright region of the CL image indicated the high free-carrier density region [8]. The stress in the dot air-bridged structure was measured by micro-Raman spectroscopy with the E_2(high) phonon mode as shown in Fig. 4(b). Different regions of micro-Raman spectroscopy were measured as in the bottom, the middle, and the top of the cross-section of the dot air-bridged sample. Compressive stress is normally observed in heteroepitaxy such as GaN/sapphire. A 4.24 cm^{-1} wavenumber blue shift of the Raman E_2(high) mode would correspond to a stress of 1 GPa. Taking the E_2(high) mode phonon frequency at 567.0 cm^{-1} as the reference value for strain-free GaN [9], the compressive stress of the dot air-bridged sample upon the interface is 0.04 GPa, which is much smaller than standard ELOG samples [10,11].

Fig. 5(a) shows the photograph of a 1.5 in-diameter, 300 μm GaN thick-film on a 2 in sapphire substrate. No crack was found either on the GaN thick-film or on the sapphire substrate. The surface of the GaN was mirror-like and the structure of the GaN/sapphire was bowed. Fig. 5(b) shows the photograph of the 1.5 in-diameter, 300-μm-thick freestanding GaN wafer, which was separated from the sapphire substrate by the laser lift-off technique with an Nd:YAG 355 nm laser. These results show that the dot air-bridged structure is a promising technique for the growth of crack-free and thick GaN films.

4. Conclusion

We have demonstrated the crack-free GaN thick-film with the dot air-bridged structure by HVPE growth. This technique can be used in relaxing the residual stress caused by different TECs and lattice constants between the GaN and the sapphire substrate. The 1.5 in-diameter, 300-μm-thick freestanding GaN wafer was fabricated by the dot air-bridged structure and the laser lift-off technique. The dislocation density was $1.8 \times 10^7\,cm^{-2}$ and the FWHM of the CL spectrum at 300 K was 77 meV, which indicated the good quality of the HVPE GaN.

In addition, the strain-reduced structure has been developed for GaN thick-film growth by HVPE. The compressive stress could be reduced to as small as 0.04 GPa to prevent the crack in the epitaxial process of GaN growth. This technique could obtain a large-area GaN thick-film, which can be used for the fabrication of freestanding GaN wafers.

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