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Spatial correlation between optical properties and defect formation in GaN thin films laterally overgrown on cone-shaped patterned sapphire substrates

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This study examines the influence of the growth mode on defect formation and strain relaxation in GaN thin films grown on cone-shaped patterned sapphire (CSPS) substrates by metal-organic chemical vapor deposition. A dramatic reduction in the luminescence dark spot density and a red-shifted excitonic emission were found for GaN on CSPS compared to GaN on planar sapphire substrates. The results also show that both the crystal quality and optical properties are improved in GaN on CSPS with a strong spatial correlation between the dark spots and local structural properties due to the induced-lateral overgrowth on the cone-shaped patterns. © *2010 American Institute of Physics*. [doi:10.1063/1.3388014]

I. INTRODUCTION

Group III-nitride wide-bandgap optoelectronic devices have been widely utilized for advanced commercial applications such as back light units in liquid crystal displays, various types of lighting fixtures, and mobile phones.[1,2] These commercial products require excellent performance from their materials, such as a high level of brightness and light emission efficiency. Consequently, a considerable amount of research has been done to enhance device performance. This includes studies of the internal quantum efficiency, light extraction efficiency, and external quantum efficiency. Among several solutions to improve the performances of the materials, the epitaxially lateral overgrowth technique is known to reduce threading dislocations effectively. However, this approach is a time-consuming process and often requires a two-step growth procedure. On the other hand, it has been known that the use of the patterned sapphire substrates (PSSs) technique can reduce the growth time because the growth occurs in a single step process. Previous PSS studies have investigated by adjusting growth parameter, such as V/III ratio, growth pressure, growth temperature, and annealing time for improving the quality of GaN thin film.[3,4] It has been reported that the PSS technique can enhance the light extraction efficiency due to optical scattering[5] and can reduce the threading dislocation density in GaN films grown on PSS.[6] However, better understanding of the defect generation and the strain relaxation is still required to optimize the lateral overgrowth mode of GaN films and to improve the performance of GaN-based optoelectronic devices.[7]

This study systematically investigates the distribution and the density of defects in GaN film grown on cone shape patterned sapphire (CSPS) and conventional planar sapphire substrates using an optical microscope, a scanning electron microscope (SEM), transmission electron microscope (TEM), high-resolution x-ray diffraction (HRXRD), and cathodoluminescence (CL) techniques. The aim of this work is to demonstrate how the induced-lateral overgrowth mode reduces the defect density and relaxes the strain in GaN thin films grown on CSPS substrates. This growth mode has also been shown to reduce edge dislocations;[8] however, there is no clear explanation of this reduction. Therefore, this study focuses particularly on the mechanism of the reduction in edge dislocations due to the induced-lateral overgrowth mode as well as the mechanism of strain relaxation by GaN on CSPS substrates.

II. EXPERIMENTS

The GaN thin films used in this work were simultaneously grown on CSPS and planar substrates by metal-organic chemical vapor deposition as shown in Figs. 1(a) and 1(b). The fabrication process of the CSPS substrate is as follows.[9] After a photoresist (PR) with a thickness of 3.5 μm was coated onto a *c*-plane (0001) sapphire substrate, the PR pattern was reflowed in a hard-baking process at 140 °C to create the shape of a cone. Subsequently, the sapphire substrate was etched using inductively coupled plasma reactive ion etching employing reactive Cl_2 gas. The diameter, height, and the interval of each cone-shaped pattern were 3 μm, 1.5 μm, and 3 μm, respectively, as shown in Figs. 1(c) and 1(d). The GaN thin films were grown on both CSPS and planar substrates under V/III ratio of 1453 for 10 min based on initial growth step. Subsequently, the growth pressure was changed from 100 to 350 Torr. The detailed growth conditions were reported elsewhere.[10] A TEM specimen was made by focused ion beam using an *in situ* liftoff method. Photoluminescence (PL) experiments were carried out using the 325 nm line excitation wavelength from a He-Cd laser with a laser power of 10 mW, in which a pho-

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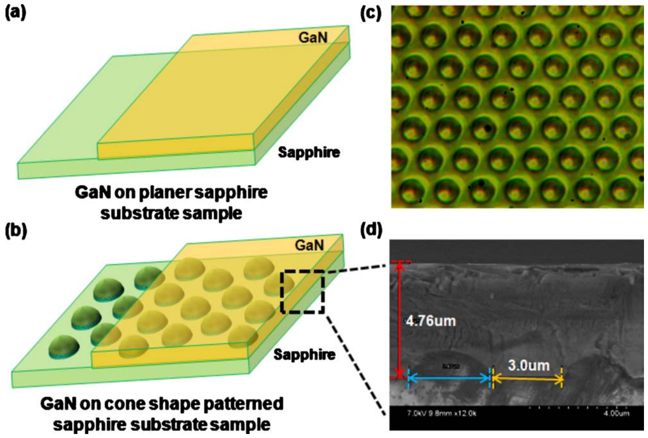

FIG. 1. (Color online) Schematics of (a) GaN on planar and (b) CSPS substrates. The diameter (3 μm), the height (1.5 μm), and the interval between each cone shape pattern (3 μm) were measured by (c) optical image and (d) cross-sectional SEM image.

tomultiplier tube was used as a detector. Time-resolved PL measurements were carried out with a frequency-doubled, mode-locked (150 fs) Ti-sapphire laser system. The PL decay curves were obtained by time-correlated single photon counting via time-to-amplitude conversions. The excitation wavelength was used 354 nm (3.5 eV) and the pulse repetition rate was controlled at 500 kHz to avoid multiple excitations.

III. RESULTS AND DISCUSSION

Figures 2(a) and 2(b) show the 10 K PL spectra and PL decay curves of GaN thin films grown on CSPS and planar substrates. In the PL spectra shown in Fig. 2(a), both the free-exciton (FX) and bound-exciton (BX) peak energies of the GaN on CSPS shifted to a lower energy by ~13.7 meV compared to the energy levels of the GaN on planar substrates. It is known that the excitonic peak energy shifts to a lower energy when the residual compressive strain is relaxed in GaN grown on sapphire substrates.[11] It was previously shown that the distance between the cones influences the value of the GaN lattice constant c (parallel to c-axis sapphire) and hence the residual strain status. A shorter distance between the cones leads to a smaller value of the lattice constant c.[12] Therefore, the presence of a cone pattern reduces the residual compressive strain in GaN due to the change of growth mode in the cone areas. Furthermore, a higher peak intensity of FX compared to BX is clearly observed for GaN on CSPS, in contrast to the case of GaN on planar substrates shown in Fig. 2(a), indicating an improvement of the optical properties for GaN on CSPS substrates.

To investigate the optical properties and carrier dynamics of recombination in GaN on CSPS and GaN on planar substrates, time-resolved PL experiments were performed at a low temperature (10 K). Figure 2(b) shows the PL decay data for GaN thin films on CSPS and planar substrates together the instrument response function (IRF) profile. It was observed that the decay time of GaN on CSPS (0.82 ns) is longer than that of GaN on planar substrates (0.28 ns). It is known that even at low temperature, the nonradiative decay rate $(1/\tau_{\mathrm{nr}})$ is dominant compared to the radiative decay rate $(1/\tau_{\mathrm{r}})$ due to high defect density in GaN thin films grown on sapphire substrates. Therefore, the observed longer lifetime of GaN on CSPS compared to that of GaN on planar substrates strongly indicates a reduction in the nonradiative centers in GaN on CSPS. From these observations, i.e., the redshift of the excitonic emission, the higher intensity of FX over BX, and the longer excitonic lifetime for GaN on CSPS, it was concluded that GaN on CSPS exhibits larger strain relaxation and better optical properties than GaN on planar substrates.

Figures 3(a) and 3(b) show symmetric (002) and asymmetric (102) reflection HRXRD ω-scan rocking curves measured for both GaN on CSPS and planar substrates, respectively. The full width at half maximum (FWHM) of the symmetry (002) ω-scan curves for GaN on CSPS (227.16 arc sec) was found to be comparable with (or slightly larger than) that of GaN on planar substrates (209.16 arc sec), whereas the FWHM of the asymmetry (102) rocking curves for GaN on CSPS was much smaller (268.32 arc sec) than that for GaN on planar substrates (323.64 arc sec). It was reported that the x-ray ω-scan curves on the symmetric (002) planes are influenced by screw-type dislocations, whereas the ω-scan curves on the asymmetric (102) planes are sensitive

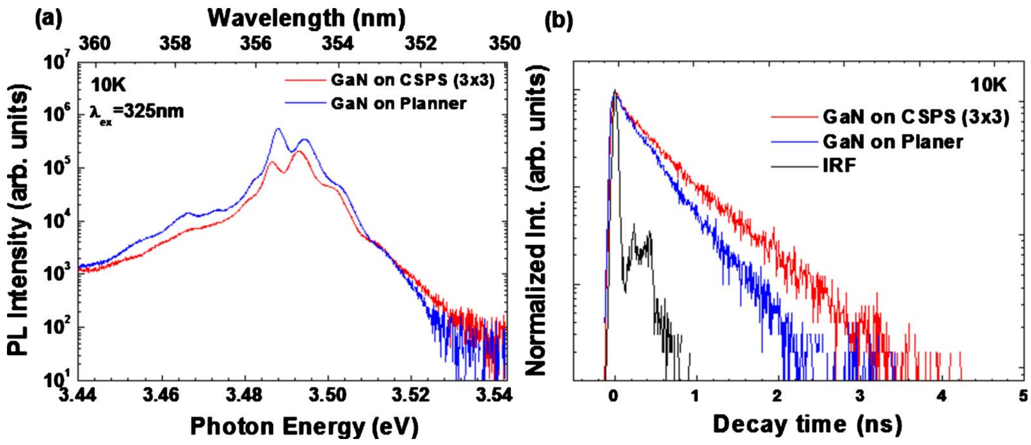

FIG. 2. (Color online) (a) 10 K PL spectra of GaN on CSPS and planar substrates. Both FX and BX peak energies of GaN on CSPS shifted to lower energy compared to those of GaN on planar substrates. (b) Temporal evolution of PL spectra at the PL peak energies of both samples and the IRF at 10 K. All spectra are normalized for clarity.

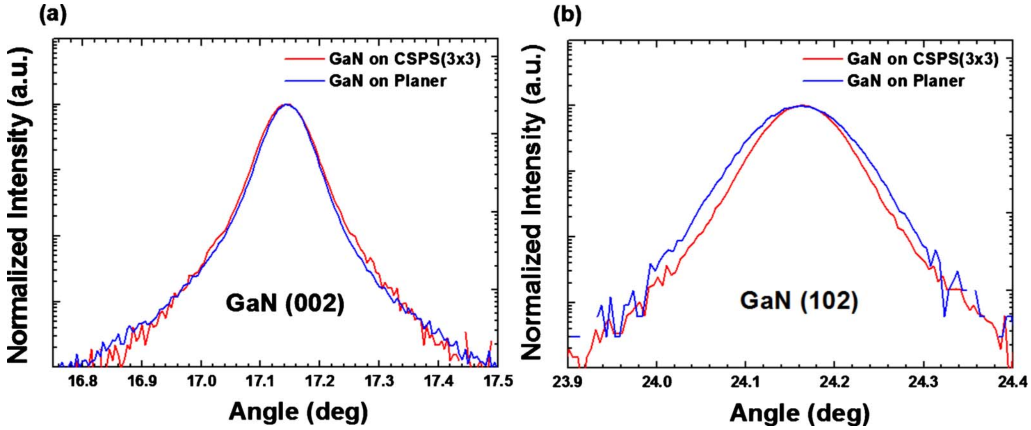

FIG. 3. (Color online) (a) Symmetric (002) and (b) asymmetric (102) reflection XRD ω-scan rocking curves measured for both GaN on CSPS and planar substrates.

to edge-type dislocations.[13] Therefore, GaN grown on CSPS shows a dramatic reduction in edge-type dislocations compared to GaN grown on a planar substrate, while the screw-type dislocations are quite similar in both cases. This can be attributed to the lateral overgrowth mode of the GaN thin film induced by the CSPS substrates.[8] In general, edge-type dislocations are easily formed in GaN thin films grown on the *c*-plane sapphire plane due to the lattice mismatch between the GaN and the sapphire. As the CSPS substrate dramatically reduces this flat *c*-plane sapphire area, the FWHM of the (102) HRXRD significantly decreases. Figures 4(a) and 4(b) show panchromatic CL images of GaN on planar and GaN on CSPS substrates, respectively. The densities of the dark spots (i.e., nonradiative recombination centers) observed in the CL images were found to be 8.0×10^8 and 9.3×10^7 cm^{-2} for GaN on planar and GaN on CSPS substrates, respectively, indicating a dramatic improvement in the crystal and optical qualities of GaN thin films due to the CSPS substrate. Interestingly, we observed a unique arrangement of the dark spot distribution on the sample surface. Directional line patterns are clearly observable, and the distance between the lines was found to be approximately 3 μm as shown in Fig. 4(b). We also compared the panchromatic CL image of GaN on the CSPS substrate [Fig. 4(c)] with an optical microscope image [Fig. 4(d)]. It was confirmed that the same area of the sample surface was imaged in both the CL and optical microscope images by selecting the same marker (indicated by the dashed ellipsoid). Subsequently, the exact location of cone- shaped pattern on CL image could be predicted by referring to the optical image. Here, the dark spots are mostly located in the area between the cones. Above the cone area, fewer dark spots are observed. A comparison of the CL and optical images verified, that the dark spots are directly located on the flat *c*-plane sapphire region of the CSPS substrate.

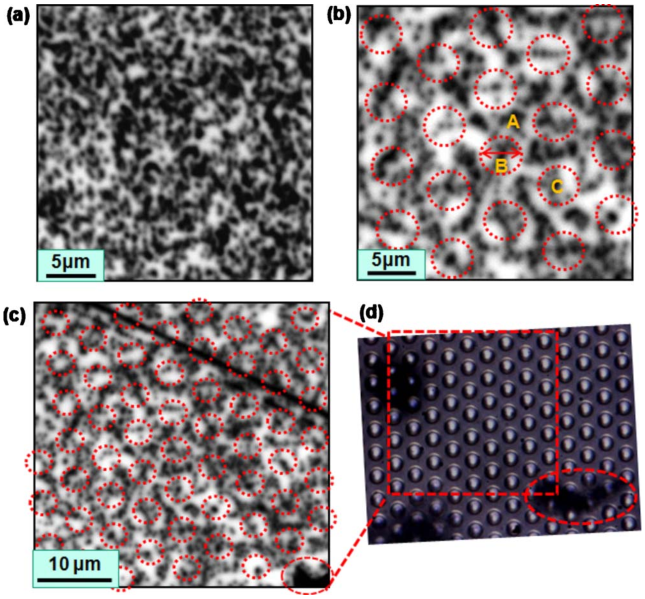

FIG. 4. (Color online) Panchromatic CL images of (a) GaN on planar and (b) GaN on CSPS substrates. (c) CL image of GaN on CSPS including target mark where the dashed lines are only guides for the eye from (d) optical image with CSPS.

In order to elucidate the relationship between the optical and structural properties and for a correlation with the lateral growth mode on the cone-shaped patterns, cross-sectional TEM experiments for GaN on CSPS were carried out, as shown in Fig. 5. It was found that the threading dislocations are mostly located on the *c*-plane region in the sapphire between the cone patterns. Based on the CL and TEM images, the GaN was categorized into the following three regions: (A) a GaN region with a high density of dark spots [Fig. 4(b)] where numerous vertical dislocations start from the flat *c*-plane sapphire area between the cone patterns [Fig. 5], as commonly observed in GaN grown on a planar sapphire substrate due to the lattice mismatch between the GaN and the *c*-plane sapphire, (B) a GaN region with few dark spots over

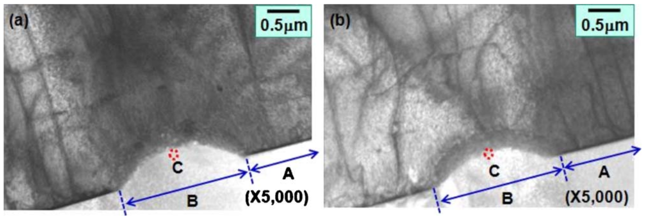

FIG. 5. (Color online) Cross-sectional TEM images of GaN on CSPS (a) without and (b) with threading dislocation on top of cone shape region.

the entire cone area [Fig. 4(b)], where there is no vertical propagation of dislocations as dislocations generated near the cone boundary bent over the cone area [Fig. 5], and (C) a GaN region with localized dark spots on the apex of the cone area [Fig. 4(b)], in which coalescence occurs at the top of the cone area from the GaN grown on the cone boundary area due to the lateral overgrowth mode [Fig. 5]. It should be noted that the bent dislocations in region B dramatically prevent the generation of threading dislocations on the surface due to the induced lateral overgrowth mode. As the growth rate of the flat area is higher than that of the cone area, the growth of the GaN on the CSPS substrate starts from the flat sapphire area between the cones. As the growth proceeds up to the top of the cone area, the laterally overgrown GaN regions become coalescent and cover the entire cone area, resulting in localized dislocations or dark spots at the apex of the cone patterns (region C). Therefore, it is concluded that the lateral overgrowth mode induced by the cone shaped pattern is responsible for the unique pattern of defect generation and for the strain relaxation in GaN thin films grown on CSPS substrates.

IV. CONCLUSION

We investigated the strong correlation between the structural and optical properties of GaN thin films laterally overgrown on cone-shaped PSSs. An improvement of the crystal quality and optical emission properties, a reduction in the residual compressive strain and a dramatic decrease in the dark spot density were observed in GaN on CSPS compared to GaN on planar sapphire substrates. We found a clear spatial correlation between the unique distribution of the dark spots shown in the CL images and the dislocations observed in TEM images. This was attributed to the lateral overgrowth mode in GaN layer grown on the cone-shaped pattern.

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