



## Regrowth of High Quality Heavily Si-Doped nGaN Utilizing Nano-Rod GaN Template

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In this study, heavily Si-doped n-type GaN (n-GaN) epitaxial layers with and without nanorod GaN (NR GaN) template were grown by metal organic chemical vapor deposition (MOCVD) system. It was found that we could achieve high-quality heavily Si-doped n-GaN layers by using the NR GaN template. It was also found that we can reduce etching pits density in n-GaN ( $1.5E19 \text{ cm}^{-3}$ ) epitaxial layer by a factor of 2.82 using the NR GaN template, as compared to the conventional sapphire substrate. © 2011 The Electrochemical Society. [DOI: 10.1149/1.3617533] All rights reserved.

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GaN-based materials have attracted much attention in recent years owing to its applications in optoelectronic devices such as light-emitting diodes (LEDs).<sup>1,2</sup> However, the performances of GaN-based LEDs grown on sapphire substrate were limited by crystal quality of the GaN epitaxial layers and current crowding effect in LED device. It is well known that crystal quality of the GaN epitaxial layers prepared on sapphire substrate is poor due to the large mismatches in lattice constant and thermal expansion coefficient between GaN and sapphire. These mismatches will result in a large number of threading dislocations (TDs) in the epitaxial layers. This could severely degrade the performance of LED device. It has been demonstrated that several methods can be used to reduce the TDs of GaN, such as epitaxial lateral overgrowth (ELO),<sup>3</sup> pendeo epitaxy (PE),<sup>4</sup> SiN or Mg<sub>x</sub>N<sub>y</sub>/GaN buffer layer<sup>5,6</sup> and patterned sapphire substrate.<sup>7</sup> Most of the methods are focused in improving undoped GaN (u-GaN) crystal quality. Moreover, it is not clear from published data how to improve Si-doped n-GaN epitaxial layer with increased SiH<sub>4</sub> flow. It is well known that crystal quality of GaN is positively correlated with the grain size of GaN. Previously, N. A. Cherkashin et al. have shown the lateral dimensions of hexagonal GaN grains decreases as increasing the Si-doped level in n-GaN.<sup>8</sup> Z.Chine et al. also have shown the tensile stress and TDs in Si-doped n-GaN increases as increasing the Si concentration in Si-doped nGaN.<sup>9</sup> In other words, TDs density increases as increasing the Si-doped level in n-GaN.

In this study, we report another possible way to improve heavily Si-doped n-GaN epitaxial layer quality is to use NR GaN template. The physical characteristic and the crystal quality of the heavily Si-doped n-GaN epitaxial layers prepared on NR GaN template will be discussed.

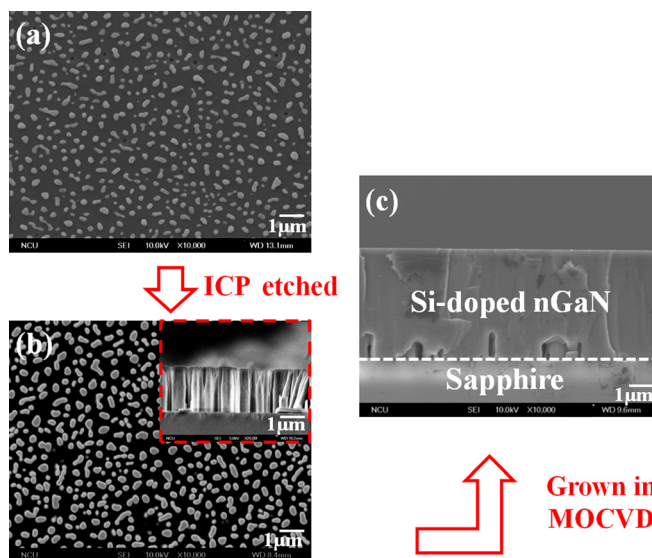
### Experimental

The NR GaN templates and the heavily Si-doped n-GaN samples used in this study were all prepared by metalorganic chemical vapor deposition (MOCVD). We first deposited a low-temperature (LT) GaN nucleation layer and a 1.8- $\mu\text{m}$ -thick undoped GaN layer on c-face sapphire substrate. A 40 nm thick SiO<sub>2</sub> film was then deposited on the GaN surface by plasma-enhanced chemical vapor deposition (PECVD), followed by deposition the 17 nm thick Ni metal film by electron-beam evaporator. The sample was subsequently rapid thermal annealing (RTA) at 800°C for 2 min to form nano-scale Ni metal islands. The self-assemble Ni nano-islands can be used as an etching hard mask, as shown in Fig. 1a. The sample was then ICP etched using self-assemble Ni nano-islands as the etching hard mask. The sample was subsequently dipped in HF solution to remove the SiO<sub>2</sub> and Ni nano-islands after etching. Figure 1b shows scanning electron microscope (SEM) image of the NR GaN templates. The fabrication process of the NR GaN templates can be found elsewhere.<sup>10</sup> It can be seen clearly that vertical GaN nanorods with an average diameter of about 250 to 500 nm were formed. It

was found the length of GaN nanorods were 1.8  $\mu\text{m}$ . It was also found that GaN nanorods density was around  $3.0 \times 10^8 \text{ cm}^{-2}$ . We subsequently deposited a heavily Si-doped n-GaN on top of the NR GaN template (i.e., NR-nGaN). We prepared NR-nGaN I, NR-nGaN II and NR-nGaN III with concentration of  $5.0 \times 10^{18}$ ,  $1.0 \times 10^{19}$  and  $1.5 \times 10^{19} \text{ cm}^{-3}$ , respectively. The thickness of NR-nGaN was 4  $\mu\text{m}$ , as shown in Fig. 1c. For comparison, a conventional heavily Si-doped n-GaN without the NR structure was also prepared (i.e., C-nGaN). The concentration of C-nGaN I, C-nGaN II and C-nGaN III were  $5.0 \times 10^{18}$ ,  $1.0 \times 10^{19}$  and  $1.5 \times 10^{19} \text{ cm}^{-3}$ , respectively. The C-nGaN consists of a 4- $\mu\text{m}$ -thick Si-doped n-GaN and a 2- $\mu\text{m}$ -thick u-GaN. The surface morphology of samples were characterized by scanning electron microscope (SEM, JEOL 7000) and atomic force microscope (AFM, Veeco D3100). The electrical properties of these n-GaN epitaxial layers were studied by Hall measurements (ACCENT HL5500) using the van der Pauw configuration in a magnetic field of 0.5 T at room temperature. The structural properties of these GaN epitaxial layers were then investigated by a double-crystal x-ray diffractometry (DCXRD, Bede D1), AFM and transmission electron microscope (TEM).

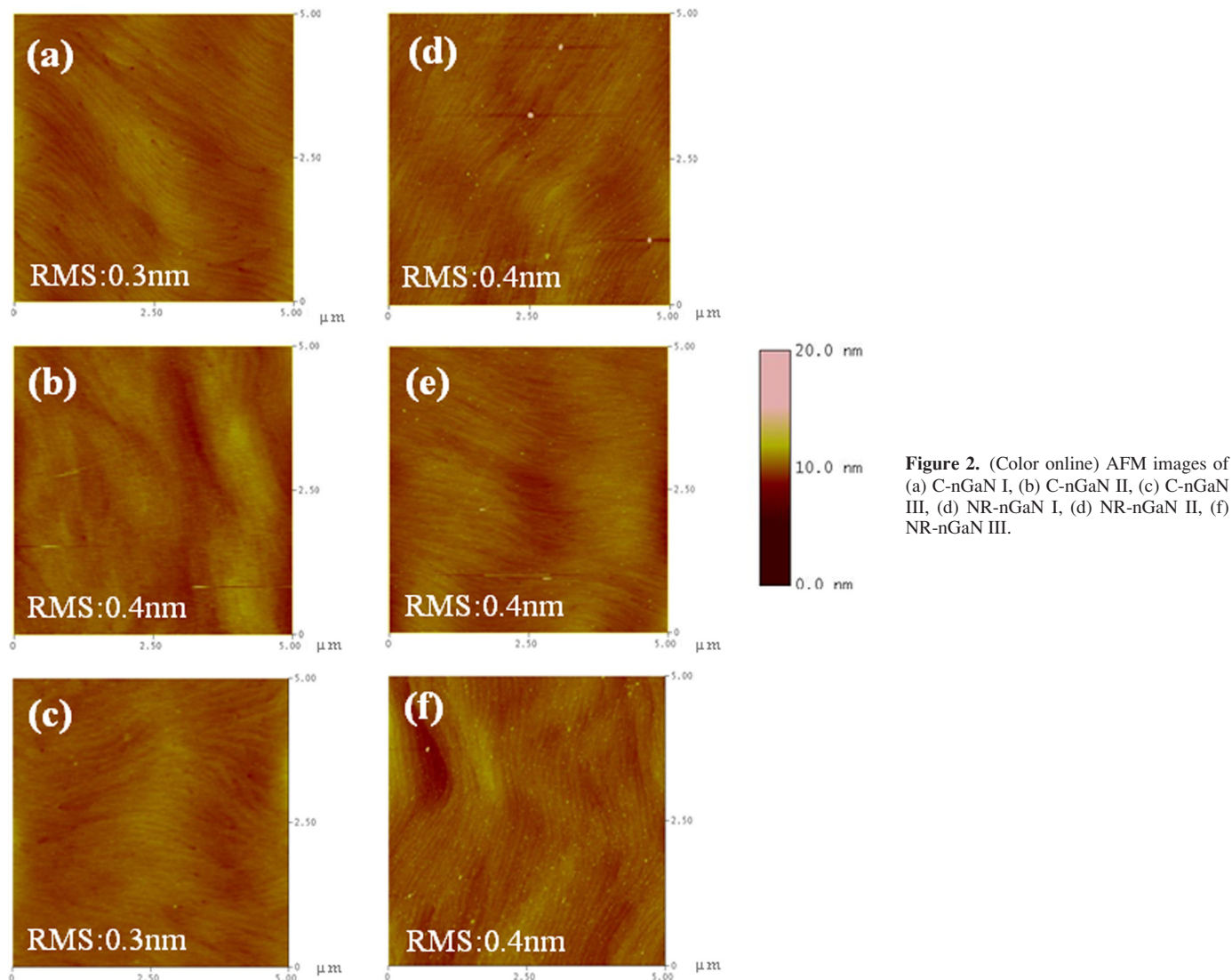
### Results and Discussion

Figure 2 shows AFM images of the Si-doped n-GaN with various carrier concentrations. It can be seen clearly that all of the sample



**Figure 1.** (Color online) SEM images of (a) Ni mask on GaN, (b) NR GaN template formed by ICP etched, (c) Si-doped n-GaN grown on the NR GaN template. Inset of Fig. 1b show the cross sectional image of NR GaN template.

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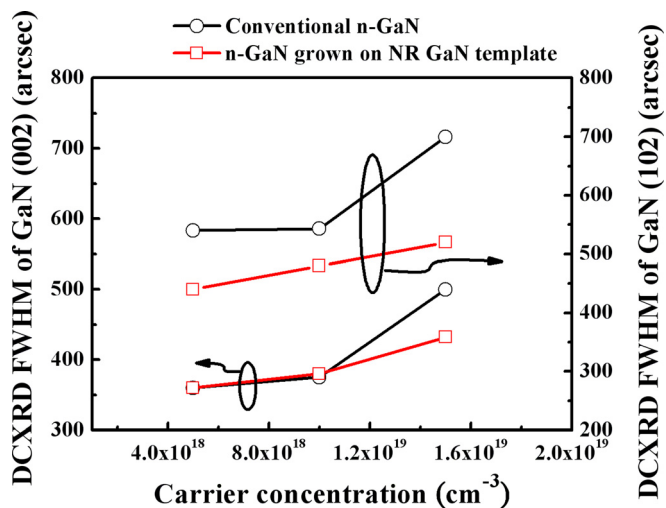


**Figure 2.** (Color online) AFM images of (a) C-nGaN I, (b) C-nGaN II, (c) C-nGaN III, (d) NR-nGaN I, (e) NR-nGaN II, (f) NR-nGaN III.

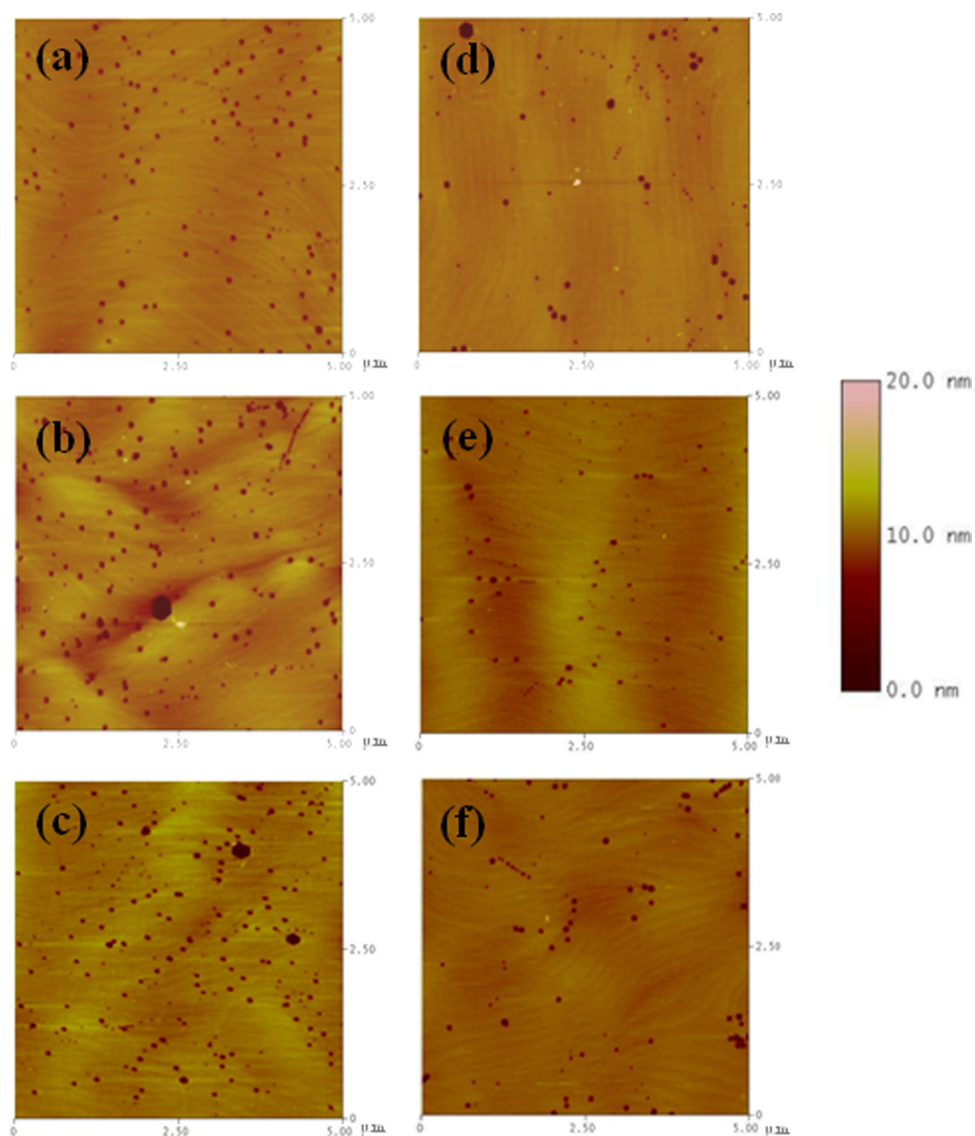
surface was very smooth. The root-mean-square (rms) roughness of Si-doped nGaNs over a  $5 \times 5 \mu\text{m}$  scanning area were 0.3–0.4 nm. In other word, high temperature n-GaN was revealed well coalesced growth on NR GaN template during lateral epitaxial overgrowth process.

Figure 3 shows the full width at half maximum (FWHM) of (0 0 2) and (1 0 2) DCXRD spectra as a function of n-GaN with various carrier concentration. In the wurtzite crystal structure, the FWHM of the (0 0 2)  $\omega$ -rocking curve is associated with the density of screw or mix dislocations, while the FWHM of the (1 0 2)  $\chi$ -rocking curve is related to all dislocations.<sup>11</sup> The FWHMs of both the (0 0 2) and the (1 0 2) rocking curves increase when the carrier concentration of n-GaN was increased. It could be seen clearly that DCXRD FWHM of the conventional n-GaN epitaxial layer was much larger than those observed from these n-GaN prepared on NR GaN template. It also should be noted that the DCXRD FWHM of both the (0 0 2) and the (1 0 2) rocking curves observed from heavily Si-doped n-GaN prepared on NR GaN template (i.e., NR-nGaN III:  $1.5 \times 10^{19} \text{ cm}^{-3}$ ) was much smaller as compared with the heavily Si-doped n-GaN prepared on conventional sapphire substrate (i.e., C-nGaN III:  $1.5 \times 10^{19} \text{ cm}^{-3}$ ). It should be attributed to the effect of lateral growth which is similar to that reported in PE<sup>4</sup> by using the NR GaN template. This finding suggests that the NR GaN template could effectively reduce the TD densities.

A wet etching experiment was conducted in  $\text{H}_3\text{PO}_4$  solution at  $250^\circ\text{C}$  to determine the etching pits density (EPD) between all six



**Figure 3.** (Color online) DCXRD FWHM of GaN (002) and (102) with various carrier concentration.



**Figure 4.** (Color online) EPD images of (a) C-nGaN I, (b) C-nGaN II, (c) C-nGaN III, (d) NR-nGaN I, (e) NR-nGaN II, (f) NR-nGaN III.

samples. These samples were examined by AFM. Figure 4 shows the EPD images over a  $5 \times 5 \mu\text{m}^2$  scanning area of n-GaN (a) C-nGaN I, (b) C-nGaN II, (c) C-nGaN III, (d) NR-nGaN I, (e) NR-nGaN II, and (f) NR-nGaN III, respectively. The EPD of n-GaN samples was  $6.7 \times 10^8$ ,  $8.4 \times 10^8$ ,  $1.1 \times 10^9$ ,  $3.6 \times 10^8$ ,  $4.2 \times 10^8$  and  $3.9 \times 10^8 \text{ cm}^{-2}$  for C-nGaN I, C-nGaN II, C-nGaN III, NR-nGaN I, NR-nGaN II, and NR-nGaN III, respectively. It was found that EPD of the these n-GaN prepared on NR GaN template was smaller than those observed from conventional n-GaN epitaxial layer. It was also found that we can reduce etching pits density in heavily Si-doped n-GaN (i.e., NR-nGaN III) epitaxial layer by a factor of 2.82 using the NR GaN template, as compared to C-nGaN III. Such a result agrees well with that observed from DCXRD results and again indicates that we could reduce the TDs of n-GaN layer by using NR GaN template.

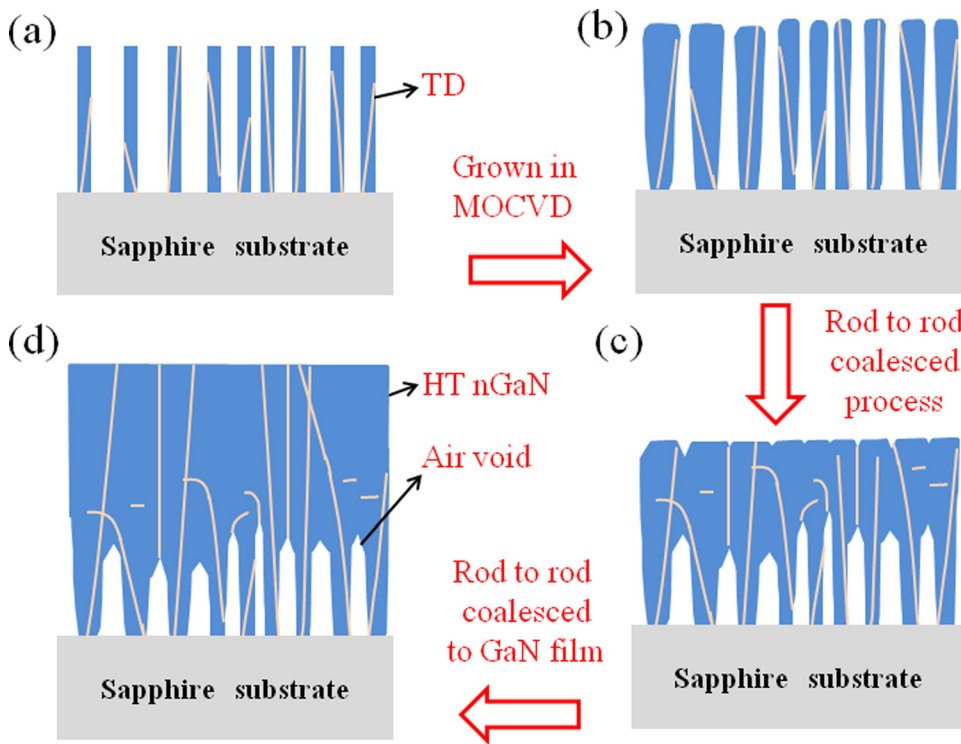
Figure 1c shows cross-sectional SEM image of the Si-doped n-GaN grown on NR GaN template. It can be observed clearly that air voids could be found at the GaN/sapphire interface.

During the growth process on NR GaN template, nano-rods grew and coalesced with each other. N-GaN epilayer could completely coalesce into a flat surface. Indeed, we found that surface of this sample is mirror-like after the growth, as shown in Figs. 2d–2f. Figure 1c shows cross-sectional SEM image of the n-GaN grown on NR GaN template. It can be seen clearly that voids could be found

at the GaN/Sapphire interface. Figure 5 shows schematic illustration of proposed growth mechanism in n-GaN on NR GaN template.

During the growing n-GaN on NR GaN template, the aspect ratio (ratio of GaN rod length to the rod-rod spacing length) could affect the gas molecule diffusion from top of GaN rods to bottom. It become more difficult for gas molecule diffuse from the top of GaN rods to the bottom when increase the aspect ratio. Such gas diffusion with different aspect ratio in chemical vapor deposition growth process has been previously modeled by Oh et al.<sup>12</sup> As the n-GaN was continued to grow in three-dimensional growth, the spacing started getting smaller from the top, as shown in the Fig. 5b. It became harder for gas molecule to diffuse down to the bottom of GaN rods. Then eventually the air voids were formed in the GaN/sapphire interface after the growing n-GaN process. Such growth mechanism on the NR GaN template maybe can promote the crystalline quality. It is well known that the TDs could have probability to bend or be terminated forming the stacking faults because of the nano scale lateral epitaxial overgrowth during the three-dimensional growth process.<sup>13</sup> Cross-sectional SEM image of Si-doped n-GaN on the NR GaN template can clearly be seen that air voids were found in the GaN/sapphire interface. The air voids may block the TDs propagation from sapphire substrate due to the large mismatches in lattice constant and thermal expansion coefficient between GaN and sapphire.





**Figure 5.** (Color online) Schematic illustration of proposed GaN growth and the proposed TDs generation in Si-doped n-GaN epitaxial layers grown on the NR GaN template.

In order to further clarify the reason for the enhancement of n-GaN quality prepared on NR GaN template, transmission electron microscopy (TEM) measurements were performed. Figure 6 are the TEM images of the NR-nGaN III and C-nGaN III, respectively. As shown in Fig. 6b, there exist a large number of dislocation in the C-nGaN III with single LT GaN buffer layer, originating from GaN/sapphire interface. It is known that generation of these dislocations is due to the large mismatches in lattice constant and thermal expansion coefficient between GaN and sapphire. In contrast, the dislocation density is much smaller for NR-nGaN III on NR GaN template, as shown in Fig. 6a. Such an observation indicates that we can effectively suppress the number of threading dislocation density by using NR GaN template due to the reduced lattice mismatch of GaN grown on NR GaN template. It was also found that a number of stacking faults occurred above the voids from NR-nGaN. It has been shown that the presence of stacking faults could block the propagation of TDs and the bending dislocation was due to the lateral epitaxial overgrowth of GaN.<sup>13</sup> Such results agrees well with that

observed from DCXRD and EPD measurement results and again indicates that we could reduce the dislocation density by using NR GaN template.

## Conclusions

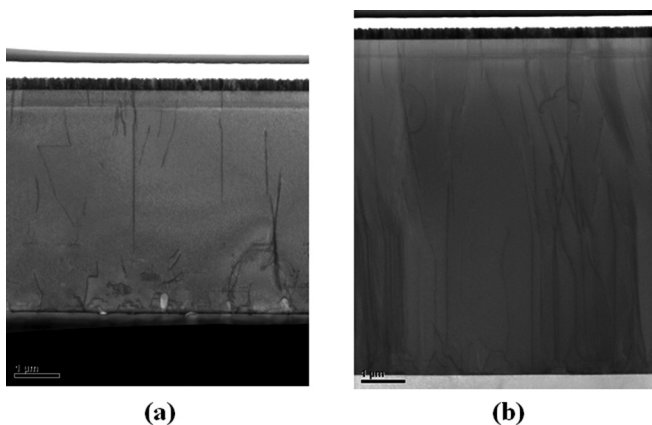
In summary, we have grown high quality and fully coalesced heavily Si-doped n-GaN epitaxial layers by using NR GaN template. It was found that we can reduce etching pits density and DCXRD FWHMs in n-GaN epitaxial layer by using the NR GaN template, as compared to the conventional sapphire substrate.

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**Figure 6.** Cross sectional TEM images of (a) NR-nGaN III, (b) C-nGaN III.