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Reduction of dislocations in GaN films on AlN/sapphire templates using CrN nanoislands

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We report significant reduction of threading dislocations in GaN films grown by hydride vapor phase epitaxy on AlN/sapphire templates by employing CrN nanoislands on the AlN. High quality GaN films with very small twist mosaic as well as small tilt mosaic have been grown on the AlN/sapphire templates, which had small tilt but very large twist mosaic. The CrN nanoislands were formed by nitridation of a thin Cr film deposited by sputtering on the AlN/sapphire template, where the AlN/sapphire template was prepared by metal organic vapor phase epitaxy. The full width at half maximum values of x-ray rocking curves from the GaN film with the CrN were 114, 209, and 243 arc sec for (0002), (10−12), and (11−20) reflections, respectively, while those of the GaN film without the CrN were 129, 1130, and 1364 arc sec, respectively. Evaluation of total dislocation density of the GaN films by plan view transmission electron microscopy revealed that the dislocation density was reduced to 2.7×10^{8} from $6.4\times10^{9}\ \mathrm{cm}^{-2}$ by employing the CrN nanoislands. The CrN nanoislands play a key role in reducing the threading dislocations by masking the propagation of dislocations as well as by bending the dislocations. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2890488]

GaN based III-nitride materials and devices have been one of the hottest issues in compound semiconductor researches and related industries. Light emitting diodes (LEDs) and laser diodes (LDs) with wavelengths in the range of blue to ultraviolet (UV) light have been commercialized based on the III-nitride materials and the industry market is continuously increasing. Performance of GaN based light emitters commonly affected by defect density and the reduction of dislocation density is highly needed. Recently, studies on high-brightness deep UV LEDs or LDs emitting at wavelengths shorter than 360 nm have been receiving increasing interests.[1,2] In order to realize high-brightness deep-UV LEDs, growth of high quality AlGaN/AlN template is important. Several methods of high quality buffers such as AlN/AlGaN superlattices, AlGaN buffer, and GaN/AlN superlattices have been reported.[3–5] Alternatively, epitaxy technique for the growth of high quality GaN on AlN template seems to be important and interesting, since if we can grow high quality GaN films on the AlN templates, we believe, the technology can be directly applied to the growth of $Al_xGa_{1-x}N$ films with any Al composition.

It has been well known that the AlN films on sapphire grown by metal organic chemical vapor deposition (MOCVD) have small full width at half maximums (FWHMs) for the symmetric (0002) x-ray rocking curve (XRC) but much larger FWHMs for the asymmetric reflections,[6–8] which means the AlN films generally have high density of edge type threading dislocations, while small density of screw type threading dislocations. Since the AlN and GaN have the same wurtzite structure, a propagation of dislocations from the AlN to the GaN films is easily expected and, therefore, protecting the propagation of threading edge dislocations from the underlying AlN into the GaN film can be a highly promising and effective way to grow high quality GaN films on the AlN/sapphire templates.

In this study, we report significant improvement of the crystal quality of GaN films on AlN/sapphire template substrates by using CrN nanoislands. Compared with the GaN films directly grown on the AlN/sapphire templates without the CrN nanoislands, very smaller FWHMs of (10−12) and (11−20) XRCs were observed for the GaN films with the CrN nanoislands. A significant reduction of edge dislocations resulting in a decrease of total dislocation density has been confirmed by transmission electron microscopy (TEM) and the possible mechanism for the dislocation reduction is discussed.

The AlN/sapphire templates with ~1 μm thick AlN were prepared by MOCVD. 15 μm thick GaN films were grown by hydride vapor phase epitaxy (HVPE) at 1040 °C on the AlN/sapphire templates with and without the CrN nanoislands. In order to form the CrN nanoislands, the Cr layer deposited by sputtering was nitrided in the HVPE reactor prior to the growth of GaN. The thickness of Cr metal layer was 4 nm and the nitridation of Cr layer was performed at around 1060 °C for 2–5 min under the NH_3 ambient. The CrN nanoislands were formed easily by such a simple nitridation process. Surface morphology and structure of the nitride Cr layer were investigated by scanning electron microscopy (SEM) and high resolution x-ray diffraction system (XRD), respectively. Crystal quality of the GaN films was addressed by measuring the XRCs for (0002), (10−12), and

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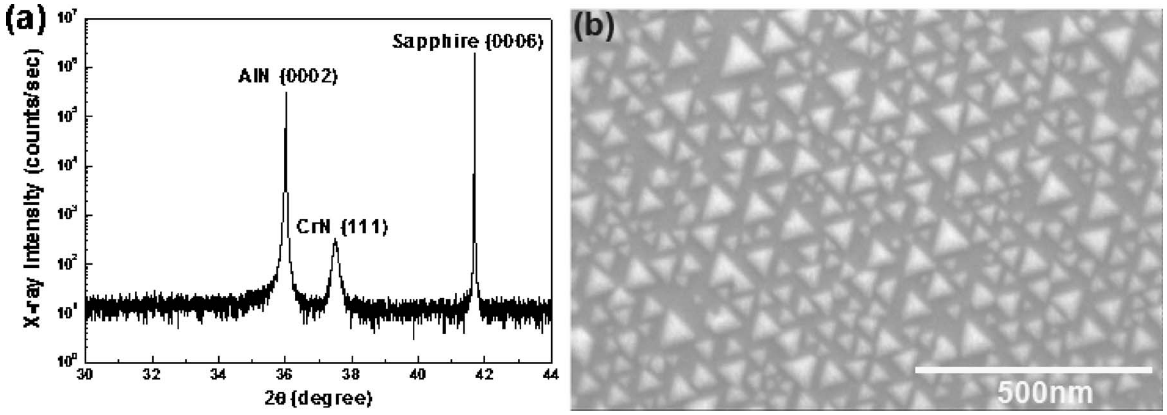

FIG. 1. XRD $\theta-2\theta$ scan (a) and SEM micrograph (b) of the Cr/AlN/sapphire sample after the nitridation.

(11−20) reflections. Dislocation structures were investigated by cross-sectional TEM observations and the dislocation density was determined by plan view observations. A TEM operating at 300 kV was used for TEM investigations.

Figure 1(a) shows a XRD θ-2θ scan of the Cr/AlN/sapphire sample after the nitridation. We can see a (111) CrN peak in addition to the (0002) AlN, which indicates the Cr layer changed to the CrN. Figure 1(b) shows a SEM micrograph of the nitrided Cr layer. As shown in Fig. 1(b), the CrN did not fully cover the AlN surface and showed tetrahedron shapes with sizes ranging from 15 to 90 nm.

Figure 2 shows the XRCs for the AlN/sapphire template and the GaN films on AlN templates with and without the CrN. As shown in Fig. 2(a), the AlN/sapphire template showed the FWHMs of 72 and 1037 arc sec for (0002) and (10−12) reflections, respectively, which is typical feature of the AlN films on sapphire substrates showing smaller and much larger FWHMs for the symmetric and asymmetric XRCs, respectively. The GaN film grown on the AlN/sapphire template without the CrN showed the FWHMs of 129, 1130, and 1364 arc sec for (0002), (10−12), and (11−20) reflections, respectively as shown in Fig. 2(b). A small increase of FWHMs of Fig. 2(b) compared with those of Fig. 2(a) indicates that the crystal quality of the GaN films mimics the AlN layer, although there is a little bit degradation of the crystal quality. Figure 2(c) shows XRCs for the GaN film on AlN templates with the CrN. The FWHMs for (0002), (10−12), and (11−20) reflections are of 114, 209, and 243 arc sec, respectively. Here, it should be noted that the FWHMs for the asymmetric XRCs were significantly decreased with sustaining the small FWHM for the symmetric XRC. This means very high quality GaN films were grown by inserting the CrN nanoislands in between the GaN and the AlN.

The dislocation density of the GaN films with and without the CrN were evaluated from the XRD measurements. The densities of screw and edge dislocations are evaluated to be 1.0×10^{8} and 8.5×10^{10} cm^{-2}, respectively, for the GaN film without the CrN, while those for the GaN with the CrN to be 7.2×10^{7} and 3.1×10^{8} cm^{-2}, respectively.

In order to confirm and get more accurate values of dislocation densities, plan view TEM observations were performed for the GaN films. Figures 3(a) and 3(b) show plan view TEM micrographs, where the pictures were taken using the method proposed by Follstaedt *et al.*,[9] where edge, mixed, and screw dislocations can be imaged from the plan view observations with the ⟨0001⟩ zone axis. The GaN film without the CrN showed a configuration of dislocations that most of dislocations with the edge type are forming low angle grain boundaries as shown in Fig. 3(a). The misaligned angle of grain boundaries was evaluated to be 0.7° on the average and the total dislocation was determined to be 6.4×10^{9} cm^{-2}. On the other hand, the GaN film with the CrN showed a significantly low density of dislocations with a

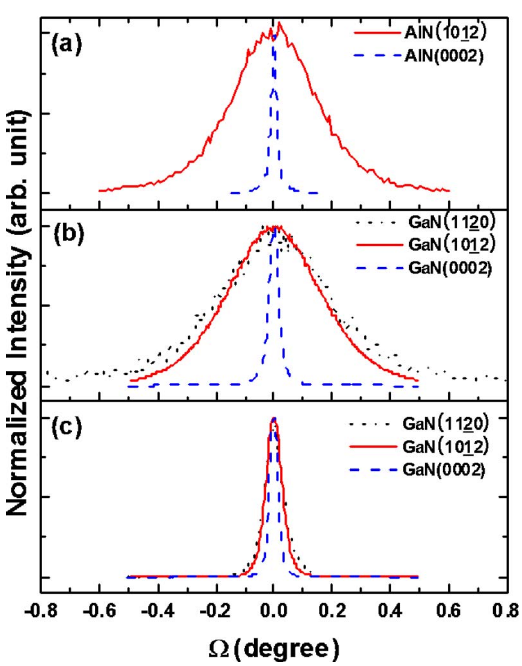

FIG. 2. (Color online) XRD XRCs for the AlN/sapphire template (a), and the GaN films on AlN templates without (b) and with (c) the CrN.

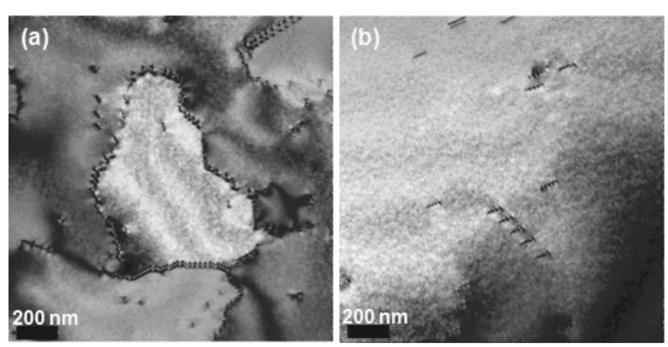

FIG. 3. Plan view bright-field TEM micrographs for the GaN films on the AlN/sapphire templates without (a) and with (b) the CrN.

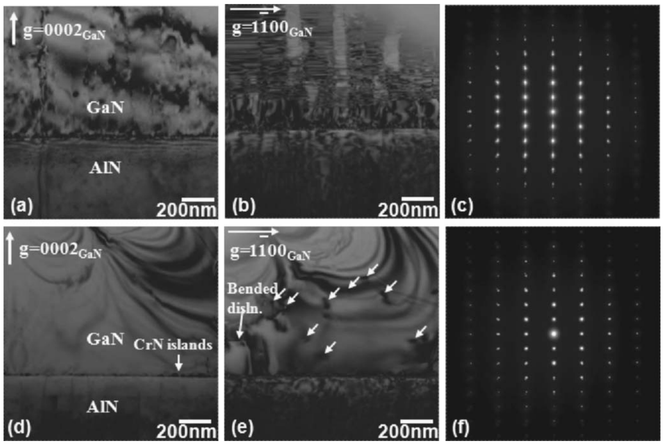

FIG. 4. Cross-sectional TEM micrographs for the GaN films on the AlN/sapphire templates without (a) and (b), and with (d) and (e) the CrN. The micrographs (a) and (d) were obtained with $\mathbf{g}=\langle 0001\rangle$, while (b) and (e) were obtained with $\mathbf{g}=\langle 1-100\rangle$. SAD patterns at the interfaces for the samples without (c) and with (f) the CrN.

configuration of random distribution, as shown in Fig. 3(b). Here, the total dislocation was determined to be $2.7\times 10^8\ \mathrm{cm}^{-2}$. The larger values of dislocation densities determined by XRD than the values determined by TEM agreed with the common tendency of overestimation of dislocation density by XRD methods compared with the TEM method.[10] Here, it should be noted that almost of the dislocations observed in our samples were edge dislocations, as shown in Fig. 3. Although the images were taken with the method of Follstaedt *et al.*, the screw and the mixed dislocations were very seldom observed because of their small numbers compared with the edge dislocations.

Figure 4 shows cross-sectional TEM micrographs and selected area diffraction (SAD) patterns at the GaN/AlN interfaces for the GaN films without and with the CrN. Figures 4(a) and 4(d) show bright field TEM micrographs under two-beam condition with excitation of $\mathbf{g}=\langle 0002\rangle$ and Figs. 4(b) and 4(e) correspond to the micrographs for two-beam condition with excitation of $\mathbf{g}=\langle 1-100\rangle$. Looking at the Figs. 4(a) and 4(b), threading dislocations in the AlN layer were mostly propagate into the GaN layer for the GaN without the CrN. Also, new dislocations were generated at the interface and thread into the GaN layer. The type of most of dislocations in the GaN film was edge. In addition, there were many basal stacking faults at about 150 nm above from the AlN/GaN interface for the GaN film without the CrN, however, no stacking faults were observed in the GaN film with the CrN. Since the stacking faults were visible in Fig. 4(b) with $\mathbf{g}=\langle 0002\rangle$ and invisible in Fig. 4(a) with $\mathbf{g}=\langle 1-100\rangle$, the type of stacking fault can be evaluated to be type II stacking fault with a displacement vector of $1/3\langle 01-10\rangle$. Such a high density of stacking faults resulted in an appearance of additional lines along the $\langle hki1\rangle$ directions in the SAD pattern in Fig. 4(c). The origin of the generation of stacking faults is not clear at current stage but we believe that the CrN nanoislands also prevent the stacking faults from generating since no stacking faults are observed in Fig. 4(e).

On the other hand, the GaN film with the CrN showed a significant reduction of edge dislocations. Most of the dislocations in the AlN template layer did not propagate into the GaN layer although some dislocations were generated at the GaN/AlN interface, which means the CrN islands did roles of masking the dislocation propagation, as shown in Figs. 4(d) and 4(e). Especially, looking at Fig. 4(e), most of threading dislocations were neither reaching to the top surface nor running along the growth direction, which was quite different from the threading dislocations in Fig. 4(b). In Fig. 4(e), we can see many short-length dislocations, as marked by arrows, and some of which looks like a so-called end-on dislocation. Considering the fact that a dislocation cannot be terminated inside the material, such images of dislocations mean that the propagation direction of dislocations was not the simple $\langle 0001\rangle$ direction but the direction was changed inside the GaN film, which resulted in the reduction of threading dislocations on the top surface region. We also see one dislocation that the propagation direction of which turns to the left and we mentioned it as a bended dislocation in Fig. 4(e). Very similar mechanisms of dislocation reductions have been reported in the MOCVD grown GaN films by using the Si_xN_y nanomask[11] or the TiN nanonetworks.[12]

In summary, we have demonstrated that the density of threading dislocations can be efficiently reduced by using the CrN nanoislands on AlN/sapphire templates. The FWHMs of the GaN film with the CrN for (0002), (10−12), and (11−20) reflections were of 114, 209, and 243 arc sec, respectively, while those of the GaN film without the CrN were 129, 1130, and 1364 arc sec, respectively. Considering the FWHMs of 72 and 1037 arc sec for (0002) and (10−12) reflections, respectively, for the AlN/sapphire template, the results obtained in this study means that we could grow the high quality GaN films with very small twist mosaic as well as small tilt mosaic on the AlN/sapphire templates, where the AlN/sapphire templates had the small tilt mosaic but very large twist mosaic. Total dislocation density of the GaN films evaluated by plan view TEM observations revealed that it reduced to 2.7×10^8 from $6.4\times 10^9\ \mathrm{cm}^{-2}$ by using the CrN nanoislands. The CrN nanoislands play a key role in reducing the threading dislocations by masking the propagation of dislocations as well as by bending the dislocations. The studies on effects of CrN nanoislands coverage, i.e., the density of nanoislands will be the next topic for investigation.

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