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Low dislocation densities and long carrier lifetimes in GaN thin films grown on a SiN_x nanonetwork

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Significant improvement of structural and optical qualities of GaN thin films on sapphire substrates was achieved by metal organic chemical vapor deposition with *in situ* SiN_x nanonetwork. Transmission electron microscope (TEM) studies revealed that screw- and edge-type dislocations were reduced to 4.4×10^7 and 1.7×10^7 cm^{-2}, respectively, for a ~5.5-μm-thick layer. Furthermore, room temperature carrier lifetimes of 2.22 and 2.49 ns were measured by time-resolved photoluminescence (TRPL) for samples containing single and double SiN_x network layers, respectively, representing a significant improvement over the previous studies. The consistent trends among the TEM, x-ray diffraction, and TRPL measurements suggest that *in situ* SiN_x network reduces line defects effectively as well as the point-defect-related nonradiative centers. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2433754]

Despite the remarkable advances achieved by III nitrides, the performances of devices are still hampered by relatively low-quality epitaxial layers grown on SiC or sapphire substrates.[1] So far, epitaxial lateral overgrowth (ELO) is the most effective method resulting in low thread dislocation (TD) densities (mid-10^6 cm^{-2}) in the wing areas.[2] However, a high concentration of defects exists in the window regions and coalescence boundaries, preventing long radiative lifetimes from being obtained. Alternative schemes, adopting the ELO concepts but using *in situ* SiN_x (Ref. 3) or *ex situ* TiN_x (Refs. 4 and 5) masks, have been reported to result in significant reduction in TDs, along with point defects. In this letter, we report on low dislocation density thin GaN films with long carrier lifetimes grown by metal organic chemical vapor deposition (MOCVD) using *in situ* SiN_x nanonetworks.

In situ SiN_x networks were formed on ~2-μm-thick GaN/Al_2O_3 wafers, followed by ~3.5 μm overgrowth of GaN at 200 Torr without any interruption. Other details of growth conditions can be found elsewhere.[6] The deposition time of SiN_x was varied from 0 min (control) to 6 min, and the effect of a second SiN_x network layer was also studied. For SiN_x deposition times of 4.5 min or less, the entire 2 in. wafer was fully coalesced, and the 5 min SiN_x sample had a fully coalesced central area (2/3 of the wafer) and a few hexagonal holes near the edge of the wafer. However, large area coalescence was not achieved in the 6 min SiN_x sample even after a ~4.5 μm overgrowth.

Table I summarizes the structural and optical properties of samples with different SiN_x deposition times. The best lifetime values achieved in a 15 min annealed TiN_x sample and a freestanding GaN from Ref. 5 are also listed for comparison. A monotonous decrease of x-ray diffraction (XRD) rocking curve full width at half maximum (FWHM) values with increasing SiN_x deposition time from 0 to 5 min was observed for both the symmetric and asymmetric reflections, indicating that both screw- and edge-type dislocations were effectively reduced by the SiN_x network. It is also noted that $(10\bar{1}2)$ FWHM values decreased more dramatically than those for the (0002) peak, suggesting a more effective reduction of edge-type dislocations by the porous network.

The cross-sectional transmission electron microscopy (TEM) image of the sample with a 5 min SiN_x is shown in Fig. 1. Most TDs were blocked by the SiN_x network, and those emanating from the pores were mostly bent into horizontal configurations. It is suggested that after the *in situ* deposition of SiN_x network, small GaN islands (seeds) are first nucleated at the pores of the SiN_x layer with $(1\bar{1}01)$ facets[7,8] (images obtained by scanning electron microscopy are not shown here), then grow vertically and laterally until a fully coalesced surface is achieved. During this evolution, dislocations are easily bent toward the facet surface. During the lateral growth stages, some dislocations may propagate laterally until they run into other dislocations at the coalescing boundaries. This mechanism has already been dubbed as the "two-step growth[9]" or the "facet-controlled ELO" (Ref. 10) for $(1\bar{1}00)$ patterned ELO to reduce TDs in both window and wing areas. In our case, we believe that naturally formed $(1\bar{1}01)$ facets play a key role in TD reduction when using the *in situ* SiN_x network, since the total reduction in TD density is achieved not only by the blocking efficiency of the SiN_x network but also by the degree of dislocation bending. The desired growth procedure, therefore, requires three major steps. The first step is the proper control of SiN_x coverage by adjusting the deposition time and or the silane flux. The second step is to promote seeds with $(1\bar{1}01)$ facets by growing

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TABLE I. XRD, TEM, and TRPL decay constants and amplitude ratios (A_2/A_1) for GaN thin films with different SiN_x deposition times.

SiN_x (min)	XRD FWHM (arc min) (0002), $(10\bar{1}2)$	TEM TDs ($\times 10^7$ cm^{-2}) edge, screw	TRPL at 200 μJ/cm^2 τ_1, τ_2 (ns)	A_2/A_1
0	4.20,6.75		0.18,0.40	0.74
3	4.10,5.76		0.20,0.52	0.66
4	3.94,5.09		0.24,0.81	0.90
4.5	3.77,4.32	9.6, 10	0.35,1.21	0.71
5	3.62,3.52	1.7,4.4	0.45,2.22	0.75
6			0.53,2.67	0.88
4+4.5	3.65,3.61		0.49,2.49	0.75
TiN_x[a]	6.43,6.11		0.47,1.86	0.59
Freestanding GaN[a]			0.34,1.73	0.33

[a]Reference 5.

at a relatively high pressure. The last step involves the enhancement of lateral growth rate by adopting a high-temperature,[10] low-pressure,[10] and Ga-rich growth condition.[11] The dislocation bending strongly depends on the GaN seed growth conditions in the second step. If the seed layers were grown at low pressures (e.g., 30 Torr), as in our earlier study on SiC substrates,[6,12] the island density would be high and uniform, and all islands would have a small height to base ratio.[10] However, at a higher pressure (e.g., 200 Torr used in this study), the seed density is relatively low, and GaN islands have a larger height to base ratio and $(1\bar{1}01)$ side facets.[6,10] As TEM studies indicated in selectively grown GaN through circular patterns, dislocations were significantly bent after the formation of pyramids with $(1\bar{1}01)$ facets.[8] On the other hand, no dislocation bending was evident from the TEM studies on our earlier samples grown at 30 Torr.[12] Therefore, between these two types of seeds discussed here, the ones grown at high pressure would bend dislocations more effectively because of the larger $(1\bar{1}01)$ facet area[8] and less island density. For the same reason, a high growth pressure is also desirable in the last step for dislocation bending, but a thicker layer is needed to achieve complete coalescence.

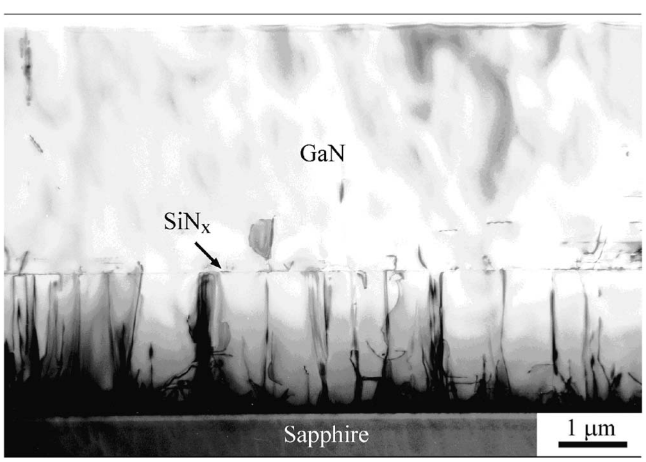

FIG. 1. Cross-sectional TEM micrograph of a GaN thin film grown with 5 min *in situ* SiN_x network by MOCVD on sapphire.

Plan view TEM images of samples with 4.5 and 5 min SiN_x (Fig. 2) reveal a much larger reduction in dislocation density than that observed in our earlier study on SiC substrates[12] or that reported by Fang *et al.*[13] Direct counting from the images indicated that the total dislocation densities were reduced to 2×10^8 and 6×10^7 cm^{-2} for the 4.5 and 5 min SiN_x samples, respectively. A small change in SiN_x

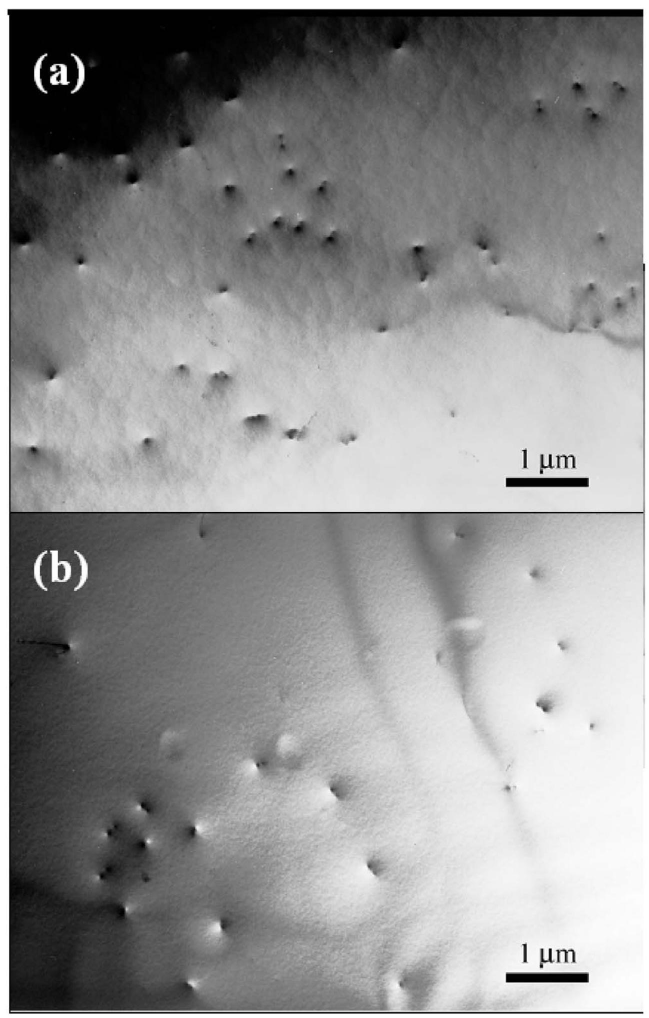

FIG. 2. Plan view TEM micrographs of GaN thin films grown with 5 min *in situ* SiN_x network by MOCVD on sapphire substrates.

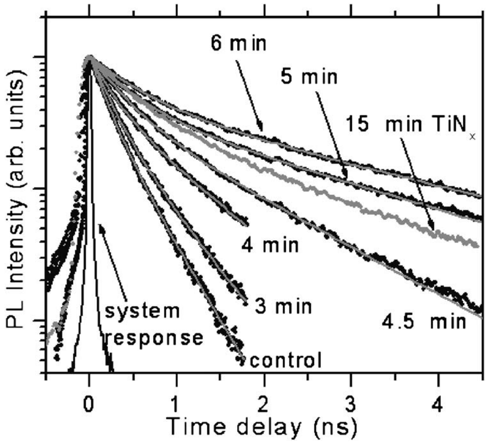

FIG. 3. Normalized time-resolved PL spectra for GaN thin films grown with different *in situ* SiN_x deposition times, control sample, and a 15 min annealed TiN_x sample from Ref. 5. The solid lines are biexponential fits to the data.

deposition time (0.5 min) resulted in a factor of 3 difference in total TD density, indicating that TD reduction is extremely sensitive to SiN_x coverage and, therefore, the seed density.

To further evaluate the effect of SiN_x network on point defect reduction, time-resolved photoluminescence (TRPL) measurements were performed using 325 nm (3.81 eV) excitation from a Ti:sapphire oscillator/regenerative amplifier pumped optical parametric amplifier. Figure 3 shows the 200 $\mu J/cm^2$ excitation TRPL data normalized to 1 for samples with different SiN_x deposition times and also for a 15 min annealed TiN_x sample from Ref. 5 for comparison. The decays for all samples were fitted by a biexponential decay function, $A_1 \exp(-t/\tau_1)+A_2 \exp(-t/\tau_2)$, and the decay constants and the amplitude ratios (A_2/A_1) obtained from the fits are summarized in Table I. The measured decay times, τ_1 and τ_2 for the fast (A_1) and slow (A_2) decaying components, respectively, are both limited by nonradiative recombination. Longer decay times and larger A_2/A_1 ratios indicate reduced nonradiative relaxation pathways. Consistent with the observations from XRD and TEM analyses, both decay times increased with increasing SiN_x deposition time. The fully coalesced 5 min SiN_x sample exhibited a slower decay and larger A_2/A_1 ratio (τ_1=0.45 ns, τ_2=2.22 ns, and A_2/A_1=0.75) as compared to the best TiN_x network sample (τ_1=0.47 ns, τ_2=1.86 ns, and A_2/A_1=0.59) reported earlier.[5] For a longer SiN_x deposition time (6 min), the surface was not fully coalesced, but the decay times nevertheless continued to lengthen (to τ_1=0.35 ns and τ_2=2.674 ns). The improvement in decay times with increasing SiN_x coverage is attributed to the reduction of threading dislocation, which in turn reduces the nonradiative recombination centers.

To check the effectiveness of multiple nanonetworks on dislocation reduction, we have prepared another ~5.5-μm-thick sample employing a second 4 min SiN_x network deposited after nominal 400 nm growth on an initial 4.5 min SiN_x network. For this fully coalesced sample XRD FWHM values (see Table I) were between 5 and 4.5 min SiN_x samples. However, the TRPL decay times (τ_1=0.49 ns and τ_2=2.49 ns) for this double SiN_x sample were longer than both single 4.5 and 5 min SiN_x network layers. Keeping in mind the correlation between XRD linewidths and density of dislocations, the increase in decay times from the single 4.5 min SiN_x sample to the double SiN_x sample might at first be attributed to the reduction in dislocation density. However, the increase from 5 min SiN_x sample to double SiN_x sample suggests that the improvement in decay times has its genesis in the reduction of nonradiative recombination centers caused by both TDs and associated point defects that cannot be detected by either TEM or XRD.

In summary, proper three-step growth process at relatively high deposition chamber pressures, as compared to previous reports on SiC substrates,[12] significantly improved the structural and optical qualities of GaN thin films grown by MOCVD. Consistent trends among XRD, TEM, and TRPL results suggest that extended defect density and nonradiative recombination center density are correlated. Remarkably long radiative lifetimes attained with SiN_x nanonetworks show that the method would be very useful in light emitting and detecting devices.

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