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Near-field scanning optical microscopy and time-resolved optical characterization of epitaxial lateral overgrown *c*-plane and *a*-plane GaN

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Epitaxial lateral overgrowth (ELO) was employed for both *c*-plane and *a*-plane GaN layers on sapphire, and a more pronounced optical improvement was observed for the *a*-plane GaN as evidenced by the significantly increased band edge photoluminescence (PL). Room temperature near-field scanning optical microscopy studies explicitly showed enhanced optical quality in the wing regions of the overgrown GaN due to reduced density of dislocations, and for the *a*-plane ELO GaN sample the wings and the windows were clearly discernible from PL mapping. Time-resolved PL measurements revealed biexponential decays with time constants that were significantly enhanced for the *a*-plane ELO GaN (τ_1=0.08 ns, τ_2=0.25 ns) when compared to the non-ELO control sample but were still much shorter than those for the *c*-plane ELO GaN (τ_1=0.26 ns, τ_2=0.90 ns). © *2006 American Institute of Physics*. [DOI: 10.1063/1.2424677]

GaN device technology has advanced considerably in the recent years owing to the vast amount of materials research focused on providing better material quality with better optical and electrical performance.[1] The device performance critically depends on the density of threading dislocations (TDs), which are inherent to epitaxial layers grown on SiC or sapphire substrates due to lattice mismatch. Among various growth techniques proposed, epitaxial lateral overgrowth (ELO) has been shown to reduce the density of TDs and associated point defects effectively and led to demonstration of GaN-based laser diodes.[2] Another characteristic associated with GaN is that most of the GaN-based devices of today employ *c*-plane oriented layers, which exhibit large spontaneous and strain induced piezoelectric polarizations which, for instance, cause spatial separation of electrons and holes in quantum wells that are used for active regions in light emitting diodes. This separation results in reduced quantum efficiency and redshift of the emission, the amount of which depends on the carrier injection level. To avoid such problems, nonpolar planes of GaN, *m* plane and *a* plane, may be employed. The *a*-plane variety can be grown on *r*-plane sapphire,[3] while the growth of *m*-plane GaN has been achieved only on *m*-SiC (Ref. 4) and γ-$LiAlO_2$,[5] which both have limited availability and higher cost compared to sapphire. However, the material quality of *a*-plane GaN layers grown on *r*-plane sapphire is still poor compared to the *c*-plane variety due to larger density of TDs and stacking faults,[6] even though some improvement has been achieved with ELO.[7,8] In this letter we report on near-field and far-field optical characterizations of both *c*-plane and *a*-plane GaN ELO samples grown by metal organic chemical vapor deposition (MOCVD) on sapphire substrates.

For the present investigation, two ELO samples were prepared by MOCVD: one with *a*-plane and the other with *c*-plane orientation. For the *a*-plane ELO sample, a 1.5-μm-thick $(11\bar{2}0)$ *a*-plane GaN template was grown on an $(1\bar{1}02)$ *r*-plane sapphire substrate with a low temperature GaN nucleation layer.[3] For the *c*-plane ELO sample, a 3-μm-thick (0001) GaN template was grown on *c*-plane sapphire with a low temperature GaN buffer layer. An approximately 100-nm-thick SiO_2 layer was deposited on both templates by remote plasma enhanced chemical vapor deposition. Using conventional photolithography and buffer oxide etch, a stripe pattern with 4-μm-wide open windows and 10-μm-wide SiO_2 stripes was generated. The stripes were oriented along the $[1\bar{1}00]$ direction of GaN that would result in the lateral growth fronts to advance along the c^+ and c^- directions in the *a*-plane sample and along the a^+ and a^- directions in the *c*-plane sample. The patterned *a*-plane template was then reloaded into the chamber for overgrowth ($\sim$13 μm) in two different stages (2 h at 1000 °C and 3 h at 1050 °C) to achieve full coalescence.[9] For the *c*-plane ELO sample, an initial vertically enhanced growth at 76 Torr for 70 min with a trimethyl Ga (TMGa) flow rate of 157 μmol/min was followed by enhanced lateral growth at 30 Torr for 4.5 h with the TMGa flow rate lowered to 118 μmol/min. The NH_3 flow rate and the temperature were kept at 7000 sccm (sccm denotes cubic centimer per minute at STP) and 1030 °C, respectively, throughout the growth. The chamber pressure was then increased back to 76 Torr for 10 min and later to 200 Torr for 10 min with other parameters kept the same to achieve a flat surface for 8 μm total overgrowth.

For evaluation of the material quality, low temperature photoluminescence (PL) was performed on the overgrown GaN layers using 325 nm excitation from a HeCd laser. The PL spectrum for the *a*-plane ELO sample at 15 K is shown in Fig. 1. The near band edge emission is composed of two peaks at 3.475 and 3.419 eV. The 3.475 eV peak, which has a full width at half maximum value of 19 meV, is a combination of the free exciton and donor bound exciton transitions, which are broad and cannot be delineated even at 15 K. The band edge emission is around two orders of magnitude stronger than that in a control sample grown directly

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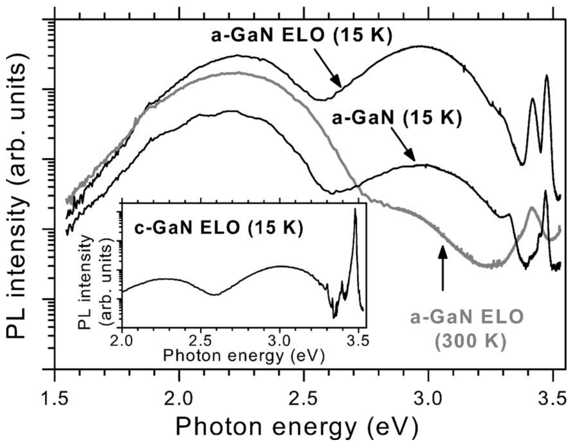

FIG. 1. PL spectra for a-plane GaN ELO sample at 15 and 300 K and for the control a-GaN sample at 15 K. Inset: PL spectra for c-plane GaN ELO sample at 15 K.

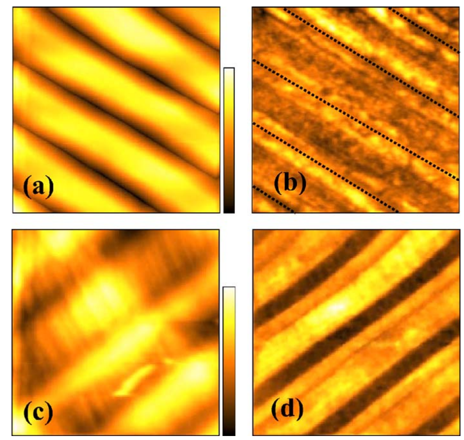

FIG. 2. (Color online) (a) AFM and (b) NSOM scans from a $50\times 50\ \mu m^2$ area of the c-plane ELO GaN sample. The dashed lines in (b) mark the meeting fronts. (c) AFM and (d) NSOM scans from a $40\times 40\ \mu m^2$ area of the a-plane ELO GaN sample. The vertical scale bars in (a) and (d) correspond to 160 and 85 nm, respectively.

on r-plane sapphire under similar conditions but without the ELO pattern (see also Fig. 1). The PL peak at 3.419 eV is most probably due to the recombination of carriers/excitons bound to stacking faults,[10] which are common in a-plane GaN. This peak has also been attributed to the recombination of excitons bound to structural defects on the surface.[11] As shown in Fig. 1, the blue band and the band edge luminescence weaken significantly at 300 K, while the intensity of the yellow band remains nearly unchanged. The inset in Fig. 1 shows the 15 K PL spectrum for the c-plane ELO GaN sample, which had a band edge emission two orders of magnitude stronger than that of the a-plane ELO GaN sample. Donor bound exciton (D^0X) and free A and B excitons (FX_A and FX_B) were clearly observed at 3.479, 3.484, and 3.492 eV, respectively, with the $n=2$ excited state of the free A exciton ($FX_A^{n=2}$) also visible at 3.503 eV. The blue and yellow emission bands were slightly weaker (four and six times, respectively) than those observed for the a-plane GaN ELO sample.

Near-field scanning optical microscopy (NSOM) measurements (contact, constant force mode) were performed using a Cryoview 2000 system (Nanonics Imaging, Ltd.) at room temperature in the illumination mode, where a 325 nm HeCd laser was used for excitation through a metal coated cantilevered optical fiber probe with 350 nm aperture. NSOM PL intensity mapping was carried out using a long working distance UV reflective objective and a photomultiplier tube to collect the overall PL spectrum with the scattered and reflected laser lights blocked by an optical filter. Figures 2(a) and 2(b) show the atomic force microscopy (AFM) and NSOM data, respectively, which were obtained simultaneously using the same NSOM fiber probe, from a $50\times 50\ \mu m^2$ region of the c-plane ELO sample. The AFM scan in Fig. 2(a) clearly shows the meeting fronts as the valleys ($\sim$100 nm deep) that are formed due to the wing tilt inherent to GaN ELO.[12] The windows (GaN seed regions) are located at the middle of two meeting fronts since the growth rates of the two opposing wings (laterally overgrown GaN regions) in c-plane ELO are the same.[12] A similar AFM image was obtained when conventional Si cantilevered probes of much smaller size (<50 nm) than the NSOM probes (>350 nm) were used. Even though the NSOM PL image in Fig. 2(b) shows somewhat localized intensity peaks most probably due to the nonuniformity of the sample, there is a significant contrast between the window and wing regions, with a stronger average PL intensity from the latter. The meeting fronts also exhibit rather lower PL intensity compared to the wings.

Figures 2(c) and 2(d) show the AFM and NSOM images, respectively, taken from a $40\times 40\ \mu m^2$ area of the a-plane ELO sample. The surface topography in Fig. 2(c) is quite different from that for the c-plane ELO GaN and shows striations along the c axis, which are common in a-plane GaN.[9] For the a-plane ELO, the meeting fronts appear as much as 60 nm higher than the windows, as also verified by AFM measurements using a conventional Si cantilevered probe (not shown), due to the miscut r-plane sapphire substrate.[9] The windows in Fig. 2(d) appear as dark regions with narrow and wide bright regions on both sides due to N and Ga polar wings (meant to indicate the polarity of the advancing growth fronts along the c^- and c^+ directions, as a-plane is nonpolar), respectively, with no significant difference between the intensities from the two. Comparison of the widths in Fig. 2(d) suggests that Ga wing is almost two times wider than the N wing, consistent with the scanning electron microscopy and transmission electron microscopy (TEM) measurements reported elsewhere.[9] Similar to the c-plane ELO sample, the meeting fronts appeared dark in the NSOM image, which may be partially due to the dislocations at these regions that were observed from TEM.[9]

In order to make sure that the correlation between the AFM and NSOM images was not an artifact related to the surface morphology and NSOM fiber geometry, reflection at 325 nm was also measured after removing the laser line-blocking filter. Considering its relatively large size, the NSOM probe may block part of the luminescence from the sample while scanning through certain surface features. The reflection data (not shown) collected from the same regions as in Figs. 2(b) and 2(d) did not provide any delineation of the wings and windows, but revealed some intensity drop at the meeting fronts. Therefore, part of the observed PL intensity drop at the meeting fronts in Fig. 2(b) may be related to this probe-size-related artifact; however, the PL intensity changes from the wing to the window regions are intrinsic to the samples. Moreover, we carefully investigated the correlations between the error signal and the optical signal to rule out any possible z-motion artifacts in the form of cross-talk

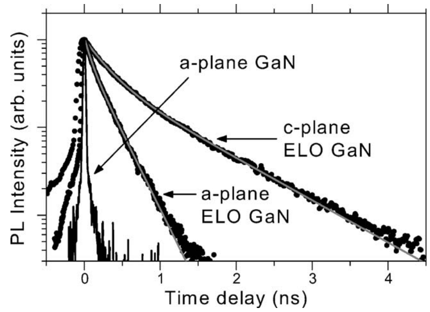

FIG. 3. Normalized room temperature time-resolved PL for *a*-plane and *c*-plane ELO GaN samples. The decay for a regular *a*-plane GaN sample is within the system resolution of ~45 ps. The solid lines represent biexponential fits to the data.

between the topographic and the optical images when the NSOM images are taken in constant force/amplitude mode.

The increased PL intensity in the wing regions of the ELO samples compared to the window regions has its origin in the improvement of the material quality in the wings by the reduction of dislocations which act as nonradiative recombination channels. Due to the nature of lateral overgrowth, no additional dislocations are generated in the wings and some of them may bend out from the windows and propagate parallel to the surface.[13] These may again bend towards the surface in the wings or counterpropagating dislocations from opposing wings may annihilate. Consequently, the TD density will be lower in the wings compared to the windows as verified by the TEM measurements,[9,13] with additional dislocations generated at the meeting fronts.[9,13] Therefore, the PL intensity is expected to be higher in the wings than in the windows and the meeting fronts.

Time-resolved PL (TRPL) measurements were also performed at room temperature using 325 nm excitation from a Ti:sapphire oscillator/regenerative amplifier pumped optical parametric amplifier and a streak camera. The excitation density was kept well below the stimulated emission threshold and the spot size (~350 μm diameter) was much larger compared to the wing/window widths. Figure 3 shows the normalized 200 $\mu J/cm^2$ excitation band edge TRPL data for the *c*-plane and *a*-plane ELO GaN samples and also for the *a*-plane control GaN sample. The decays for all the samples were well characterized by a biexponential decay function, $A_1 \exp(-t/\tau_1)+A_2 \exp(-t/\tau_2)$, which produced the fitting parameters summarized in Table I. Both of the measured decay times, τ_1 (short) and τ_2 (long), are limited by nonradiative recombination, and longer decay times and larger A_2/A_1 ratios indicate reduced nonradiative relaxation pathways.

The *c*-plane ELO sample exhibited much longer decay times (τ_1=0.26 ns, τ_2=0.90 ns) compared to the *a*-plane ELO sample (τ_1=0.08 ns, τ_2=0.25 ns), but a smaller A_2/A_1 ratio (0.60 in *c* plane compared to 1.63 in *a* plane). Faster decay times observed for the *a*-plane variety are consistent with the low temperature PL data discussed above that showed significantly lower band edge intensities for the *a*-plane samples. The decay constants for the *c*-plane ELO sample are longer than those for a control GaN sample with no ELO template (τ_1=0.18 ns, τ_2=0.40 ns) (Ref. 14) but shorter than the values reported for GaN grown on SiN_x nanonetwork templates by MOCVD (τ_1=0.49 ns, τ_2=2.49 ns) (Ref. 14) both measured under the same conditions reported here. Although shorter than those for the *c*-plane ELO sample and the single decay time of 0.43 ns reported for *a*-plane ELO GaN at much higher excitation densities (0.8 mJ/cm^2),[15] the decay times for the *a*-plane ELO sample are significantly improved compared to the control *a*-plane GaN (see Fig. 3), which exhibited a decay faster than the system response (~40 ps). The improvement in decay times with ELO of both *c*-plane and *a*-plane GaN also suggests the reduction of nonradiative channels formed by the TDs.

TABLE I. Room temperature TRPL decay constants and amplitudes for the *c*-plane and *a*-plane ELO GaN samples.

Sample	A_1	τ_1	A_2	τ_2
c-plane ELO GaN	0.59±0.01	0.26±0.01	0.41±0.01	0.90±0.01
a-plane ELO GaN	0.38±0.01	0.08±0.01	0.62±0.01	0.25±0.01

In summary, PL, NSOM, and TRPL measurements were performed for the investigation of *c*-plane and *a*-plane ELO GaN samples. From NSOM measurements, the wing regions were found to exhibit more intense luminescence than the window regions due to reduced number of dislocations in the wings. The NSOM PL contrast between the windows and the wings were not as large in the *c*-plane sample as it was for the *a*-plane ELO sample, most probably due to already much better material quality of the *c*-plane layers as verified by the PL measurements. The *c*-plane ELO sample exhibited a much longer decay (τ_2=0.90 ns) compared to the *a*-plane ELO sample (τ_2=0.25 ns) and an *a*-plane control sample with no ELO pattern (τ<0.04 ns). The results indicate that significant improvement in the material quality has been achieved with the ELO process.

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