

# InGaN/GaN multi-quantum well and LED growth on wafer-bonded sapphire-on-polycrystalline AlN substrates by metalorganic chemical vapor deposition

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## Abstract

We report growth of InGaN/GaN multi-quantum well (MQW) and LED structures on a novel composite substrate designed to eliminate the coefficient of thermal expansion (CTE) mismatch problems which impact GaN growth on bulk sapphire. To form the composite substrate, a thin sapphire layer is wafer-bonded to a polycrystalline aluminum nitride (P-AlN) support substrate. The sapphire layer provides the epitaxial template for the growth; however, the thermo-mechanical properties of the composite substrate are determined by the P-AlN. Using these substrates, thermal stresses associated with temperature changes during growth should be reduced an order of magnitude compared to films grown on bulk sapphire, based on published CTE data. In order to test the suitability of the substrates for GaN LED growth, test structures were grown by metalorganic chemical vapor deposition (MOCVD) using standard process conditions for GaN growth on sapphire. Bulk sapphire substrates were included as control samples in all growth runs. *In situ* reflectance monitoring was used to compare the growth dynamics for the different substrates. The material quality of the films as judged by X-ray diffraction (XRD), photoluminescence and transmission electron microscopy (TEM) was similar for the composite substrate and the sapphire control samples.

Electroluminescence was obtained from the LED structure grown on a P-AlN composite substrate, with a similar peak wavelength and peak width to the control samples. XRD and Raman spectroscopy results confirm that the residual strain in GaN films grown on the composite substrates is dramatically reduced compared to growth on bulk sapphire substrates.

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## 1. Introduction

Owing to the lack of low cost, large-diameter (2-in or greater) GaN substrates, GaN epitaxy is performed almost exclusively on non-lattice matched substrates [1]. For commercial InGaN/GaN LED devices the overwhelming

substrate of choice is *c*-plane sapphire [2]. Growth techniques such as low temperature nucleation layers (NLs) [3] have been developed to improve the deposition of GaN films on sapphire resulting in adequate material for many devices despite the large differences in lattice constant and coefficient of thermal expansion (CTE). Continuing issues remain, in particular the CTE mismatch is known to cause film stress and wafer bowing during growth of InGaN/GaN LED structures on sapphire [4],

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Table 1  
Comparison of thermal properties of GaN substrate materials

Material	CTE (20–1100 °C) (ppm/K) <sup>a</sup>	Thermal conductivity (W m <sup>-1</sup> K <sup>-1</sup> )
<i>c</i> -Plane GaN	5.2	210 Ref. [1]
<i>c</i> -Plane sapphire	8.1	23 Ref. [1]
P-AlN	5.5	180 <sup>b</sup>

<sup>a</sup>In-plane CTE averaged over the temperature range indicated, using data from Ref. [7].

<sup>b</sup>CeramTec North America Corp. Material Selection Guide, 2003.

leading for example to within-wafer and run-to-run wavelength non-uniformity. Strategies incorporating *in situ* curvature monitoring have been implemented on 2-in sapphire to improve the wavelength uniformity [4,5], however, even with such growth controls in place it is not possible to eliminate the CTE mismatch effects entirely. For example the issue of high residual stress upon cool-down [6] remains, which can impact the yield of post growth processing steps. Finally, in order to reduce production costs it will be important to use larger diameter substrates, however, the effects of wafer bowing on growth and processing are expected to be amplified and more difficult to control as the sapphire wafer diameter increases.

Here we present results of metalorganic chemical vapor deposition (MOCVD) growth of InGaN/GaN MQW (multi-quantum well) and LED structures on a composite substrate consisting of a thin laminate of *c*-plane sapphire attached to a polycrystalline aluminum nitride (P-AlN) support substrate. The sapphire layer provides an epitaxial template for the GaN growth, while the thermo-mechanical properties are controlled by the thick P-AlN substrate. As shown in Table 1, polycrystalline AlN has a CTE mismatch with *c*-plane GaN that is an order of magnitude smaller than the CTE difference between GaN and sapphire. The thermal conductivity of P-AlN is also approximately five times higher than that of sapphire. It is therefore expected that substrate bowing attributed to thermal gradients across the thickness of the sapphire substrate [5] as well as the CTE mismatch induced bowing experienced during growth on sapphire, can be substantially eliminated with these composite substrates. The reduced substrate bow and the higher thermal conductivity should result in improved temperature uniformity. Therefore using this composite substrate approach it should be possible to grow GaN LED's with good wavelength uniformity on a larger diameter substrate than is currently practical with bulk sapphire. Another advantage of the composite substrate is that the P-AlN support substrate is rapidly etched in hot KOH [8], so that epitaxial lift-off for flip chip bonded LED devices may be accomplished using a batch wet chemical process rather than laser lift-off techniques.

MQW and LED growth and device results for the composite substrates are compared to those obtained on

traditional bulk sapphire substrates in the same growth run. Although previous work has reported GaN growth on composite substrates, for example silicon carbide-on-insulator substrates [9], this is the first report of MOCVD III-nitride device growth on a composite substrate engineered to have a close CTE-match with GaN.

## 2. Experimental details

The composite substrates are fabricated using wafer bonding and layer transfer techniques [10] and consist of a thin layer of *c*-plane sapphire bonded to a 500 μm thick, 50 mm diameter tape-cast P-AlN substrate. Prior to bonding, a SiO<sub>2</sub> bonding layer is deposited onto the P-AlN substrate by plasma-enhanced chemical vapor deposition (PECVD) using TEOS precursor. The PECVD oxide is densified in nitrogen at 1100 °C to remove hydrogen and is then polished to a sub-nanometer RMS surface roughness and a final thickness of approximately 1 μm. Fifty millimeter diameter, double-side polished (DSP) on-axis ( $\pm 0.3^\circ$ ) *c*-plane sapphire substrates are co-implanted with H and He ions to facilitate layer transfer. The bonding and layer-transfer process is performed in a vacuum wafer bonder, and following transfer the composite substrate is annealed in nitrogen at 1100 °C to stabilize the transferred layer, which is typically about 1 μm thick. A CMP treatment is performed to reduce the surface roughness, after which the substrates are ready for growth.

The InGaN/GaN MQW and LED growths were performed in a Veeco D-125 MOCVD system. The films were grown simultaneously on single-side polished (SSP) and DSP sapphire, and the composite P-AlN/sapphire substrates. No modifications to the standard two-step growth conditions [3] were made for the composite substrates. The MQW test structure consists of a ~2.5 μm thick Si-doped n-type GaN layer grown at 1050 °C followed by a 200 nm 2% InGaN layer grown at 790 °C, and a 5-period InGaN/GaN MQW grown at 770 °C. X-ray diffraction (XRD) analysis of the MQW indicates an InGaN well thickness and indium content of 2.5 nm and 12% indium, respectively, and a GaN barrier thickness of 9.0 nm. The 2% InGaN layer, which was added to enhance the PL intensity from the MQW test structure, is not included in the LED device growth. In addition to the n-type buffer layer and 5-period MQW, the LED structure includes a 30 nm thick ~15% p-AlGaIn layer after the MQW and a 200 nm thick p-type GaN contact layer grown at 970 °C. The well thickness and indium content for the LED growth were 2.5 nm and 13%, respectively, and the GaN barrier thickness was 8.3 nm according to XRD.

## 3. Results and discussion

Structural characterization was performed on the substrate before and after GaN growth. A RHEED image of the starting sapphire template layer is shown in Fig. 1(a).

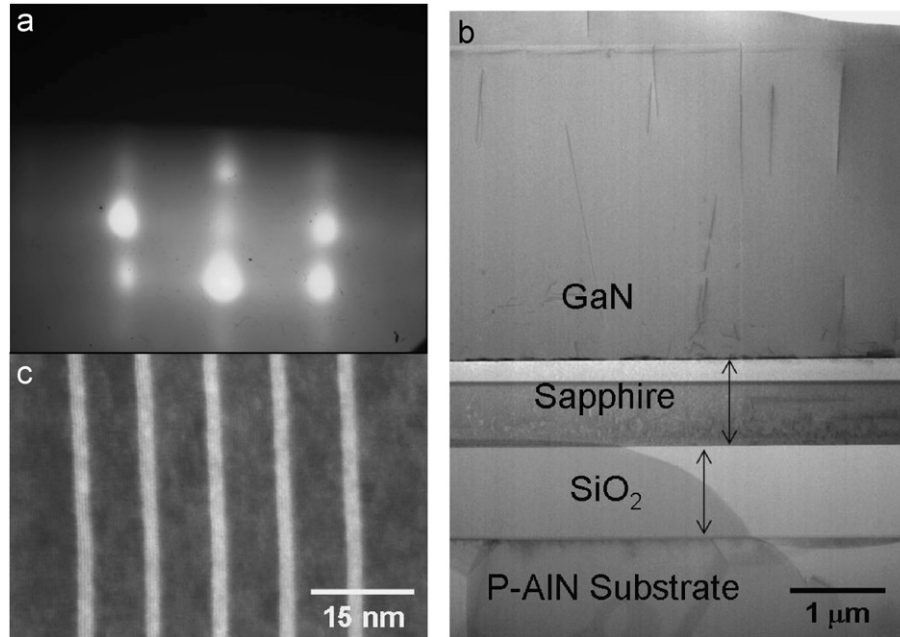


Fig. 1. (a) RHEED of transferred sapphire film on a P-AlN substrate, (b) cross-sectional TEM of GaN LED structure deposited on a sapphire/P-AlN composite substrate, and (c) higher magnification TEM of InGaN/GaN MQW region of the film in (b).

The streaky pattern indicates a relatively smooth starting surface, which we have also measured by AFM to have an RMS roughness of less than 2 nm over a  $50 \times 50 \mu\text{m}$  scan. A TEM cross-section of the composite substrate following growth of the full LED structure is shown in Fig. 1(b). The P-AlN substrate, SiO<sub>2</sub> bonding layer, sapphire template layer and GaN film are easily distinguished in this overview cross-section. The erosion of the top surface visible on the right side of the image is damage induced during the FIB processing of this TEM sample, and the contrast within the SiO<sub>2</sub> layer is an artifact of thickness variation in the foil. The contrast in the sapphire template layer (about 900 nm thick in this sample) is not clearly understood, but is possibly related to strain effects associated with the implantation process. This contrast was not observed in other samples in which the template layer was thinned to below 500 nm. As shown in the micrograph, the interfaces between the oxide bonding layer, sapphire layer, and grown GaN film remain intact during the temperature cycling associated with the MOCVD growth process. Based on the relatively low defect level we observe in this TEM foil (about 100 nm thick) and the photoluminescence (PL) and XRD results we discuss below, we judge that the GaN growth on the composite substrate is of high quality. The MQW region is visible near the top of the GaN layer in Fig. 1(b), and is shown in the higher resolution TEM image in Fig. 1(c).

The MQW test structure was grown simultaneously on a DSP sapphire, SSP sapphire, and P-AlN composite substrate. The growth was interrupted after the completion of the GaN buffer layer to enable inspection of the substrates prior to continuing the growth. *In situ* reflectance measurements obtained during growth of the

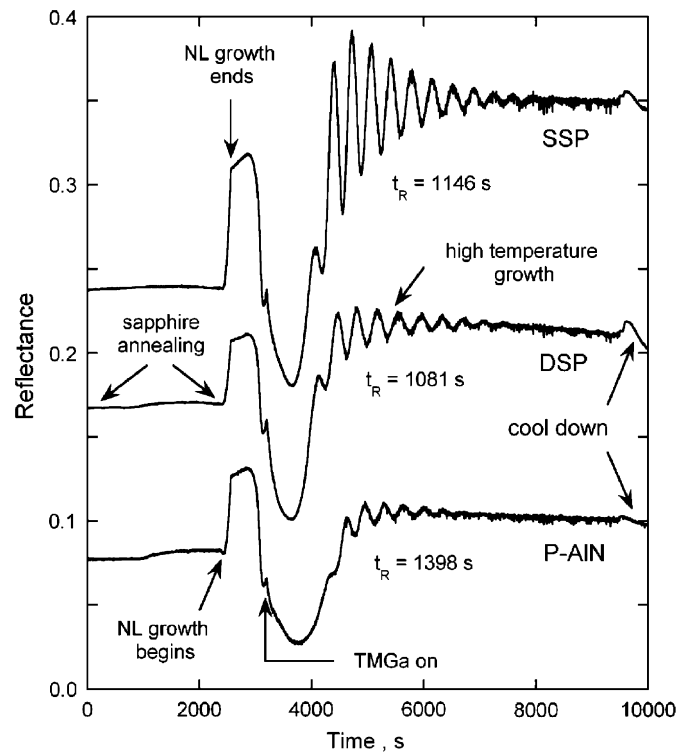


Fig. 2. Optical reflectance waveforms measured during buffer layer growth on a sapphire/P-AlN composite substrate and two bulk sapphire substrates (SSP and DSP), offset vertically for clarity.

Si-doped GaN buffer layer are shown in Fig. 2. The curves are offset vertically in the figure and important growth stages are denoted. In comparing the reflectance waveforms slight differences are observed; however, more similarities are observed than differences. For instance,

during the NL growth at 530 °C the reflectance signal increases until the TMGa is shut off. Following this, the temperature is ramped up to 1050 °C during which time the reflectance signal first increases and then decreases due to NL decomposition and recrystallization [11]. Once the TMGa is turned on again the GaN layer initially roughens and the reflectance decreases. Continued GaN growth leads to a smoothing of the film and a recovery in the reflectance signal [12]. The recovery time,  $t_R$  as defined elsewhere [11] is shown for each of the substrates. Based on the reflectance oscillations during the buffer layer growth the growth rate is determined to be approximately 1.3  $\mu\text{m}/\text{h}$  for all the samples. After the growth oscillations have subsided the SSP sapphire sample reflectance signal approaches  $\sim 0.19$  (based on a starting reference of 0.0774 for sapphire), close to that of bulk GaN. Consistent with our previous experience the increase in the reflectance is less for the DSP sapphire sample, which is attributed to a constant background signal for the DSP wafer associated with reflection from the polished backside. The increase is also less for the composite substrate, presumably due to reflections from the bonded sapphire/P-AlN interface. Backside and interface reflections similarly affect the amplitude of the growth oscillations for the DSP sapphire and P-AlN composite substrate. In summary, apart from the difference in the amplitude of the reflectance signal changes, which is expected owing to the different interface and backside reflections for the three substrates, the reflectance waveforms are very similar. The recovery time for the composite substrate ( $t_R = 1398$  s) is slightly longer than for the sapphire control substrates ( $t_R \approx 1100$  s), which could be caused by differences in the GaN nucleation dynamics associated with the different surface preparation of the composite substrate.

Following the buffer layer growth, the three samples were reloaded into the MOCVD system to continue the MQW structure. PL measurements were obtained from the MQW samples after growth and are shown in Fig. 3.

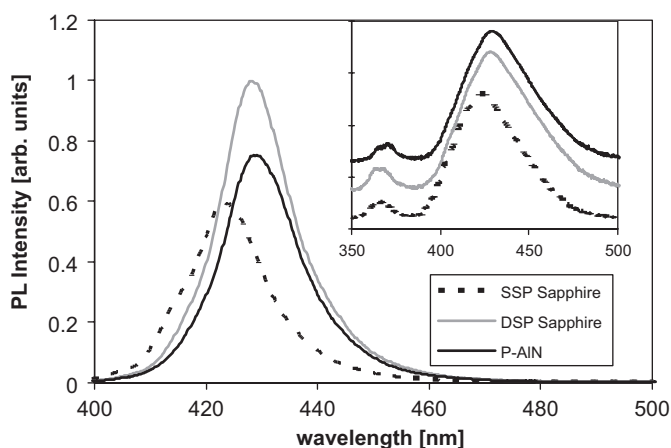


Fig. 3. Photoluminescence spectra of the InGaN/GaN MQW structure grown on bulk sapphire and composite P-AlN substrates. PL curves are scaled by the same factor and are presented on a log scale in the inset (shifted vertically for clarity).

Measurements were made using a HeCd laser at 325 nm. The PL emission wavelength from the MQW (429 nm for the DSP sapphire and composite substrate and 422 nm for the SSP sapphire sample) and peak width (FWHM of 16.1, 15.2, and 16.1 nm for the composite substrate, DSP sapphire and SSP sapphire) are similar for the films deposited on the composite substrate and the bulk sapphire control substrates. The PL intensities are also the same within a factor of two. Given the different backside surfaces and interfaces of the three substrates, we cannot make precise comparisons of film quality based on the PL intensity. We note, however, that the results appear consistent with a similar film quality for all three substrates, with a higher intensity for the DSP wafer compared to the SSP wafer as expected due to backside reflections, and an intermediate intensity for the composite substrate.

To further investigate the material quality of the MQW, high-resolution triple-axis XRD ( $\omega-2\theta$ ) scans were obtained along the (0002) direction and are shown in Fig. 4. The scans for the composite substrate and the sapphire control substrates are qualitatively similar, indicating that the material quality of the MQW growth is similar. Due to uncertainty in the XRD calibration for these scans, the absolute strain is not certain from this measurement, however, the relative peak separations are meaningful. Based on lower resolution scans (not shown) we have obtained on a calibrated tool the composite substrate sample was unstrained within  $\pm 0.03\%$ . The relative peak shift for the sapphire control samples in Fig. 4, and in scans of the full LED structure growth (not shown), corresponds to a  $c$ -axis strain of  $0.2 \pm 0.05\%$  for the sapphire substrates. The compressive strain observed in the sapphire control samples is expected due to the CTE mismatch. For isotropic biaxial strain the  $c$ -axis

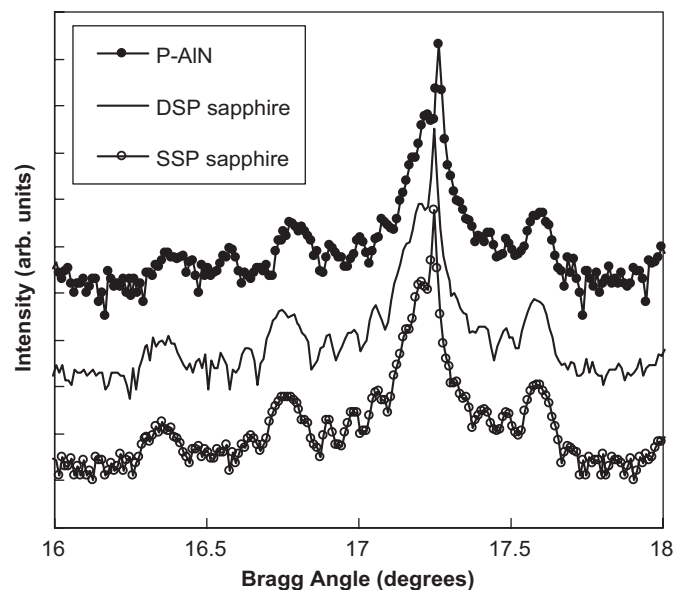


Fig. 4. (0002) XRD scans of the InGaN/GaN MQW structure. Scans are on a log scale and shifted vertically for clarity.

strain  $\varepsilon_z$  and the in-plane strain  $\varepsilon_x$  are related by  $\varepsilon_z = \varepsilon_x (v-1)/2v$  where we use a Poisson ratio of  $v = 0.2$  for GaN. This implies that the samples grown on bulk sapphire had a residual in-plane strain of  $-0.4 \pm 0.1\%$  (compressive). Using the CTE values in Table 1 the in-plane strain on the sapphire control samples associated with cool-down from  $1050^\circ\text{C}$  is expected to be  $-0.3\%$ , in fair agreement with the residual strain measured by XRD.

In order to corroborate the XRD measurement of the post-growth film strain we have performed Raman spectroscopy measurements. The results are summarized in Table 2. The peak shift  $\Delta\omega$  of the  $E_2$ -high phonon mode of GaN is related to the in-plane biaxial stress by  $\sigma_x = k\Delta\omega$  where  $k$ , the pressure coefficient for GaN is  $2.9\text{ cm}^{-1}\text{ GPa}^{-1}$  [13]. As a reference for the unstrained peak position we measured an HVPE-grown free-standing GaN substrate, for which the peak was at  $567.1 \pm 0.1\text{ cm}^{-1}$ . The peak positions for the film grown on the DSP sapphire substrate and composite substrate were  $569.8$  and  $567.3\text{ cm}^{-1}$ , respectively. This corresponds to an in-plane stress of  $930\text{ MPa}$  (compressive) for the film grown on sapphire compared to  $70\text{ MPa}$  for the composite substrate. Using the relation for the in-plane strain  $\varepsilon_x = (1-v) E^{-1} \sigma_x$  and a value of  $E = 300\text{ GPa}$  for the Young's modulus of GaN, we calculate an in-plane strain of  $\varepsilon_x = -0.25 \pm 0.02\%$  for the growth on the sapphire substrate in good agreement with the predicted in-plane CTE strain. Consistent with the

XRD result of no measurable strain for the film grown on the composite substrate, the strain calculated from the Raman measurement for this sample is  $-0.02 \pm 0.02\%$ . This is also consistent with the CTE values in Table 1 which would predict a strain of  $-0.03\%$  for the composite substrate growth. The value of the residual strain in the GaN films also depends on the intrinsic strain during growth, however, these results are consistent with the GaN film being essentially relaxed prior to cool-down on both the sapphire and composite substrates.

The full LED structure was grown on three new substrates in a single growth run. Cross-sectional TEM images of the grown layers following completion of the LED growth were obtained using on-zone imaging conditions, and are shown in Fig. 5(a) and (b) for the DSP sapphire and composite substrate, respectively. Visually the density of dislocations is lower in the film grown on the composite substrate by a factor of about two. In order to facilitate more quantitative comparisons of the images, the thickness of each TEM foil was determined by SEM imaging, and is shown schematically in Fig. 5. Based on the dislocations visible in the region  $500\text{ nm}$  immediately below the MQW, where the thickness of each foil is  $\sim 100\text{ nm}$ , the dislocation density in the DSP sapphire sample is  $2.3 \times 10^9\text{ cm}^{-2}$  and that of the composite substrate sample is  $6 \times 10^8\text{ cm}^{-2}$ . More extensive measurements are required to determine how typical these dislocation density estimates are over the substrates; however, these measurements indicate that the LED film growth on the composite substrate is at least as high quality as that grown on the sapphire control wafer. A defect structure is visible in the transferred sapphire layer of the composite substrate in Fig. 5(b) (black arrow) and extends to the GaN/sapphire interface. We note, however, that no dislocations are observed in the GaN material directly above the defect. This implies that small defects in the sapphire template resulting for example from the implantation, layer-transfer,

Table 2  
Raman spectroscopy measurements of in-plane GaN film stress

	HVPE GaN reference	GaN on DSP sapphire	GaN on composite substrate
$E_2$ -high phonon peak ( $\text{cm}^{-1}$ )	567.1	569.8	567.3
In-plane stress (MPa)	–	930	70
In-plane strain (%)	–	–0.25	–0.02

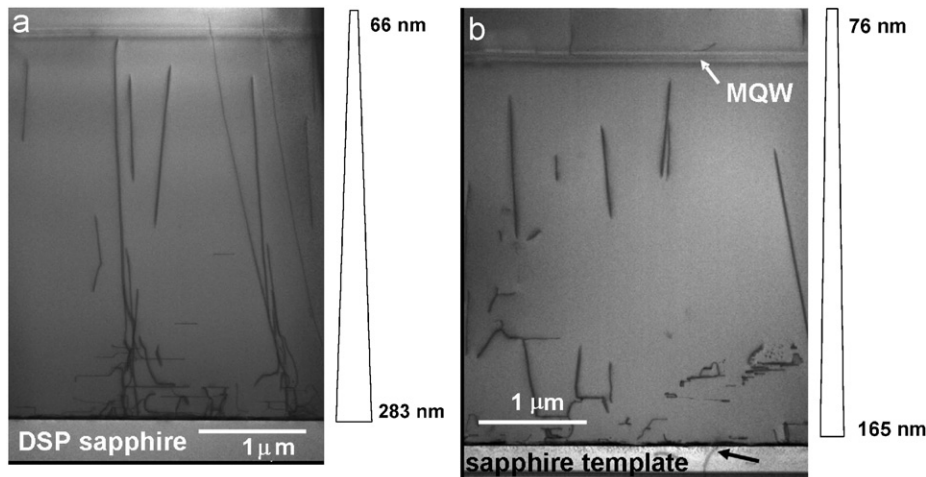


Fig. 5. Cross-sectional TEM images of LED structure grown on: (a) DSP sapphire and (b) sapphire/P-AlN composite substrate. Foil thickness as determined by SEM is shown at the right of each image.

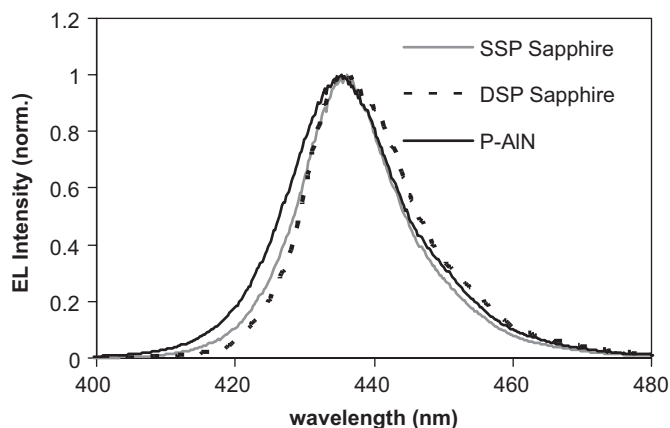


Fig. 6. Normalized electroluminescence spectra of LED structures grown on a composite sapphire/P-AlN substrate and bulk sapphire substrates.

or CMP process do not necessarily propagate into the epitaxial GaN layer.

A simple test configuration was used to measure the on-wafer electroluminescence (EL) of the LED structure grown on all three samples. For the EL measurements simple contacts are made to the n- and p-sides and the intensity measured at constant current. Fig. 6 shows the normalized EL spectra obtained from the center of each of the three substrates. The peak wavelength (436 nm) is essentially the same ( $\pm 1$  nm) for all the samples. The peak widths are also similar (FWHM of 16, 17 and 18 nm, respectively, for the SSP sapphire, DSP sapphire and composite substrate). Because the blue light emission is measured through the substrate, the difference in transmission characteristics of the three substrates (reflections from the backside surface or from internal bonding interfaces, and internal scattering and absorption in the P-AlN) precludes a direct comparison of the EL intensities. As a rough test to account for the transmission loss through the P-AlN substrate, we inserted a bare P-AlN substrate (nominally identical to the one used to fabricate the composite substrate) between the SSP sapphire sample and the detector. Using this P-AlN ‘filter’ we find that the EL intensity from the SSP sapphire sample is the same within a factor of two of the intensity measured from the composite substrate sample. In order to properly compare the EL efficiencies it will be necessary to make fully processed flip-chip devices.

#### 4. Conclusion

A novel composite substrate for GaN LED growth has been demonstrated which is designed to eliminate the CTE mismatch effects commonly experienced with growth on sapphire substrates. MQW and LED test structures grown on the composite substrates using standard recipes had similar material quality and emission wavelength as those grown on sapphire control substrates in the same run, according to TEM, XRD, PL and EL measurements. *In situ* reflectance measurements during the buffer layer

growth also showed similar behavior for the composite substrate and bulk sapphire. We conclude that the 2-step growth process on the composite substrates proceeds in a similar way to growth on sapphire. As anticipated, the films grown on the composite substrates had substantially reduced residual strain compared to the sapphire control substrates, as measured by both XRD and Raman spectroscopy. The magnitude of the strain difference after cool-down is in agreement with published CTE data, and indicates that the composite substrates are nearly CTE-matched to GaN. Cross sectional TEM measurements indicate a dislocation density for the composite substrate LED growth that is at least as low as that on a sapphire substrate. Further work is required to investigate how the reduction in CTE mismatch stress affects the growth uniformity and post-growth processing characteristics of these substrates compared to bulk sapphire.

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