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Structural and optical quality of GaN grown on $\text{Sc}_2\text{O}_3/\text{Y}_2\text{O}_3/\text{Si}(111)$

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Thick (~ 900 nm) GaN layers were grown by molecular beam epitaxy on cost-effective $\text{Sc}_2\text{O}_3/\text{Y}_2\text{O}_3/\text{Si}(111)$ substrates and characterized by x-ray diffraction and photoluminescence. Samples grown in Ga-rich condition show superior structural and optical quality with reduced density of cubic GaN inclusions within the hexagonal matrix and a relatively strong photoluminescence emission at 3.45 eV at 10 K. Cubic inclusions are formed in the initial growth stage and their concentration is reduced with increasing film thickness and after rapid thermal annealing. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.3699201>]

I. INTRODUCTION

Optoelectronic and electronic applications of GaN rely mostly on high quality layers grown on sapphire and silicon carbide substrates. These expensive substrates constitute a major fraction of the final GaN-based device cost, inhibiting the widespread adoption of GaN technologies, for example, GaN-based light emitting diodes (LED) in solid state lighting. To reduce costs, new methods for GaN growth on large size Si wafers are under development. Buffer layers are used to overcome the challenges of GaN epitaxy on Si (55% thermal and 17% lattice mismatch, interfacial reactions etc.) including AlN,¹ HfN,² ZnO,³ and bixbyite oxides.^{4,5} Epitaxial bixbyite oxides on Si with a wide variety of physical properties can serve as a platform for overgrowth by high quality semiconductors, such as Si and Ge.⁶ Recently, we presented a lattice engineered $\text{Sc}_2\text{O}_3/\text{Y}_2\text{O}_3$ heterostructure as an approach to integrate single crystalline, wurtzite GaN on Si(111).⁵ The oxide buffers can be also utilized not only to form the active layer of GaN on Si but also to deposit distributed Bragg reflectors (DBR), which improve light extraction efficiency and reduce optical losses resulting from the absorption by Si substrate.^{7,8} Here, we report on the application of the $\text{Sc}_2\text{O}_3/\text{Y}_2\text{O}_3$ buffer to grow thick (~ 900 nm) GaN layers on Si(111) and the comprehensive GaN films characterization. The GaN layers were deposited in either N- or Ga-rich regimes, close to the stoichiometric condition. We show that Ga-rich regime results in superior structural and optical GaN quality with reduced density of cubic inclusions and strong photoluminescence at 3.45 eV.

II. EXPERIMENTAL

$\text{Sc}_2\text{O}_3/\text{Y}_2\text{O}_3$ heterostructures were prepared in a multi-chamber molecular beam epitaxy (MBE) system on 4 in. Si(111).⁵ After oxide deposition, wafers were transferred

in situ for GaN overgrowth. Two sets of GaN samples were prepared. First set was grown in the N-rich regime (second in Ga-rich) with the substrate temperature, Ga beam equivalent pressure (BEP), N flow and power of radio frequency plasma source of 720 °C, 2×10^{-7} mbar, 0.4 sccm, and 300 W, respectively (720 °C, 5×10^{-7} mbar, 1 sccm, and 300 W, respectively). GaN growth was studied *in situ* by reflection high energy electron diffraction (RHEED) and GaN surface topography by scanning electron microscope (SEM). X-ray diffraction (XRD) was carried out using Cu K α radiation in line focus geometry (9 kW rotating anode). The threading screw (TSD) and threading edge dislocation (TED) densities in the GaN films were calculated based on a kinematical XRD approach.⁹ Chemical interdiffusion was investigated by energy-dispersive x-ray spectroscopy (EDX) in a scanning transmission electron microscope (STEM). For STEM images a high angle annular dark field (HAADF) detector was used. Optical properties were studied by photoluminescence (PL) (He-Cd laser, $\lambda = 325$ nm).

III. RESULTS AND DISCUSSION

Figure 1 summarizes RHEED and corresponding SEM results for two representative samples from each set. A spotty RHEED pattern [Fig. 1(a)] obtained after growth of a 900 nm GaN layer in the N-rich regime (low Ga/N ratio) is the result of a faceted surface visible by SEM in Fig. 1(b). For an increased Ga/N ratio (Ga-rich regime), spots in the RHEED are replaced by streaks [Fig. 1(c)]. The 3×3 GaN surface reconstruction (white arrows) indicates that this 800 nm GaN film is N-polar.¹⁰ SEM of this sample [Fig. 1(d)] reveals that the GaN layer is composed of coalesced blocks (grains) with a smooth surface.

High resolution XRD analysis for the 800 nm film grown in the Ga-rich regime are displayed in Fig. 2. The sample structure is illustrated by a high resolution TEM image in Fig. 2(a). The specular θ - 2θ scan [Fig. 2(b)] reveals the vertical growth orientation, namely GaN(0001)/ $\text{Sc}_2\text{O}_3/\text{Si}(111)$.

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FIG. 1. (a) RHEED and (b) SEM of 900 nm GaN film grown in N-rich regime. (c) RHEED and (d) SEM of 800 nm GaN film grown in Ga-rich regime. RHEED patterns were recorded with e-beam aligned along GaN $\langle 11\bar{2} \rangle$ azimuth.

$\text{Y}_2\text{O}_3(111)/\text{Si}(111)$. TSD and TED densities were estimated by measuring the GaN(0004) [Fig. 2(c)] and $(10\bar{1}0)$ [Fig. 2(d)] rocking curves, respectively. Initial evaluation of the GaN crystalline perfection is obtained from the full width at half maximum (FWHM) of the rocking curves, which is 0.39° for the GaN(0004) and 0.55° for the GaN($10\bar{1}0$) peak. More detailed analysis was accomplished by fitting the diffraction peaks.⁹ Best fits are displayed as red curves in

Figs. 2(c) and 2(d) and reveal TSD and TED densities of 3.1×10^{10} and $2.6 \times 10^{10} \text{ cm}^{-2}$, respectively. Similar analysis performed for the 900 nm sample grown in the N-rich regime gives TSD density of 9.1×10^{10} and TED density of $3.3 \times 10^{11} \text{ cm}^{-2}$.

The crystalline quality of the GaN layers grown on $\text{Sc}_2\text{O}_3/\text{Y}_2\text{O}_3/\text{Si}(111)$ is dependent on the GaN layer thickness and post deposition rapid thermal annealing (RTA). This behavior is similar to that observed for growth on sapphire.¹¹ Figure 3 summarizes the influence of the GaN layer thickness and RTA (N_2 ambient) on the FWHM of the GaN(0004) [Fig. 3(a)] and $(10\bar{1}0)$ [Fig. 3(b)] peaks. For samples grown in the N-rich regime the FWHM of the GaN(0004) peak [Fig. 3(a)] decreases from 1.5° for a 150 nm film to 0.7° for a 900 nm layer. This reduction results probably from a gradual annihilation of TSDs with increasing thickness. Further GaN(0004) width reduction by about 12% independent of the GaN thickness can be achieved by RTA at 900°C . Similar trends are observed for samples grown in the Ga-rich regime; the FWHM decreases from 0.5° for a 450 nm sample to 0.4° for a 800 nm layer. Also in this case, the GaN(0004) peak narrows after RTA by about 23%. Changes in the FWHM of the GaN($10\bar{1}0$) reflection are shown in Fig. 3(b). Samples grown under N-rich conditions show FWHM of 2.5° at 150 nm decreasing to 1.8° at 900 nm. Samples grown under Ga-rich regime show FWHM of 0.6° at 450 nm and 0.5° at 800 nm. Figure 3 gives thus evidence that Ga-rich growth condition result in smaller FWHMs of both

FIG. 2. (a) TEM cross section of the GaN/ $\text{Sc}_2\text{O}_3/\text{Y}_2\text{O}_3/\text{Si}(111)$ heterostructure; viewed along a $\langle 11\bar{2} \rangle$ azimuth; (b) specular θ - 2θ scan; rocking curve scan over (c) GaN(0004) and (d) GaN($10\bar{1}0$) reflection; fitting curves shown by red lines.

FIG. 3. FWHM of GaN (a) (0004) and (b) (10 $\bar{1}$ 0) peaks as a function of thickness and growth conditions.

GaN(0004) and (10 $\bar{1}$ 0) rocking curves, implying better crystalline quality in comparison with N-rich conditions. Additionally, post deposition RTA has a significant influence on the GaN(0004) peak width for samples grown in both regimes. In contrast, the same RTA treatment has negligible impact on the FWHM of the GaN(10 $\bar{1}$ 0) reflection.

In situ RHEED study of the initial GaN growth stage reveals the co-existence of (111) oriented cubic (cub-GaN) and (0001) oriented hexagonal (hex-GaN) domains (data not shown). As growth proceeds, intensity of the RHEED pattern associated with the cub-GaN gradually decreases and disappears when the GaN film thickness reaches about 20 – 30 nm. To investigate the relation between the cub-GaN and the hex-GaN phases as a function of GaN thickness, ex-situ XRD pole figure studies were performed ($2\theta = 70^\circ$).¹² An example is shown in Fig. 4(a) for a 900 nm GaN layer grown on Sc₂O₃/Y₂O₃/Si(111) under N-rich conditions. The presence of the diffraction spots originating from Si and the buffer (labeled in Fig. 4(a)) indicates that the complete GaN layer is measured. The appearance of the hex-GaN{20 $\bar{2}$ 1}

and cub-GaN{311} reflections at χ angles of 75° (green circle) and 29° (red circle) proves the presence of both hex and cub phases in the film. For clarity, ϕ scans along the green and red circles are extracted and plotted in Fig. 4(b). It is seen that the intensity of the hex-GaN{20 $\bar{2}$ 1} reflections is about three times higher than that of the cub-GaN{311}. In addition, the six GaN(311) peaks indicate twinned GaN cubic inclusions.⁶ Figure 4(c) compares the intensity ratio of the cub-GaN(311) and hex-GaN{20 $\bar{2}$ 1} reflections for different GaN layer thicknesses and growth conditions. It is noted, that due to different structure factors and scattering geometries of the investigated diffraction peaks, the intensity ratio is not identical to the cub-GaN/hex-GaN domain population ratio. In general, as-grown GaN layers prepared in the N-rich regime show higher cub/hex intensity ratio than films grown in the Ga-rich regime at comparable thicknesses. The ratio is also clearly dependent on the GaN layer thickness. For samples grown in N-rich regime, the cub/hex intensity ratio decreases from 0.3 for 30 nm layers to 0.036 for 900 nm films. For samples grown in the Ga-rich regime the ratio decreases from 0.029 at 470 nm to 0.011 at 800 nm. These results corroborate the *in situ* RHEED observations and prove that the cub-GaN inclusions are formed in the initial growth stage at the GaN/Sc₂O₃ interface and their formation is suppressed during further GaN growth. Probably, cub-Sc₂O₃(111) acts as nucleation seed for cub-GaN(111). Furthermore, the concentration of cub-GaN in the hexagonal matrix is higher for samples grown in N-rich regime than for those grown in Ga-rich regime. This behavior might be attributed to an interfacial reaction between the Sc₂O₃ and the impinging N and Ga atoms and the formation of N-O bonds in the interface region during growth. Such scenario was previously proposed to explain formation of cubic inclusions in hex GaN grown on ZnO and Al₂O₃.¹³

The cub/hex intensity ratio can be influenced by RTA for samples prepared in both growth regimes (Fig. 4(c)). For example, for a 800 nm layer grown in Ga-rich regime the cub/hex ratio is reduced from 0.011 to 0.0006 after RTA at 900°C , suggesting that cub-GaN is partially transformed into hex-GaN. While the origin of the cub-GaN formation is still under investigation for the GaN/Sc₂O₃/Y₂O₃/Si(111), it can be stated that, based on EDX, it is not directly related to oxygen diffusion into the GaN layer, as previously suggested.¹⁵ Normalized concentration profiles of Si, O, Y, Sc, N, and Ga were determined by EDX analysis before and after RTA of a 600 nm GaN layer grown in the N-rich regime

FIG. 4. (a) Pole figure ($2\theta = 70^\circ$) of 900 nm GaN grown in N-rich regime on Sc₂O₃/Y₂O₃/Si(111); (b) Φ scan across hex-GaN{20 $\bar{2}$ 1} at $\chi = 75^\circ$ and cub-GaN{311} at $\chi = 29^\circ$; (c) cub-GaN(311)/hex-GaN(20 $\bar{2}$ 1) intensity ratio as a function of thickness; (d) EDX profile of the heterostructure.

FIG. 5. Cross-section HAADF STEM images of GaN/Sc₂O₃/Y₂O₃/Si(111) heterostructure before (left) and after RTA (right).

FIG. 6. Low temperature (10 K) PL spectra from thick GaN films grown in the N-rich (black) and Ga-rich regime (red).

(Fig. 4(d), profiles of Sc, Y, and N are not shown). The oxygen profile at the GaN/Sc₂O₃ interface is identical for the GaN layer before and after RTA. In addition, after RTA, formation of amorphous Y-silicate at the Y₂O₃/Si interface is observed.¹⁴ The existence of around 5 nm-thick amorphous Y-silicate layer after RTA is confirmed by a cross-section HAADF images presented in Fig. 5. Note that the Si substrate is partially consumed by the interfacial Y-silicate layer. No further RTA-induced changes in the oxide buffer layer structure and the GaN/Sc₂O₃ interface are detected.

Figure 6 shows 10 K PL spectra from the GaN films grown in N- and Ga-rich regimes. The spectrum for the 800 nm GaN layer grown under Ga-rich conditions is dominated by a relatively sharp (FWHM of 22 meV) and relatively intense emission peak at 3.45 eV (donor bound exciton transition (D⁰X)).¹⁶ Yellow luminescence (YL) usually detected at 2.22 eV is not clearly distinguishable for this sample. On the contrary, GaN samples grown under N-rich condition (900 nm GaN film) exhibit broad defect-related YL and clear peaks at 3.42 eV (D⁰X, FWHM of 77 meV) and 3.33 eV (probably due to overlapping emission spectra related to longitudinal optical phonon replica of the D⁰X line, donor-acceptor pair transition, and excitons bound to structural defects). Shift in the D⁰X line position between N-rich and Ga-rich samples is associated with the different donor/impurities incorporated in the samples grown in two different regimes.

IV. CONCLUSIONS

In summary, we investigated the growth of thick GaN layers on Sc₂O₃/Y₂O₃/Si(111) substrates. The GaN films were deposited in two growth regimes: N-rich and Ga-rich, close to the stoichiometric condition. In the initial GaN growth stage, the co-existence of cubic and hexagonal GaN domains is observed. As growth proceeds, the formation of cubic GaN inclusions is suppressed and hexagonal GaN layers on Sc₂O₃/Y₂O₃/Si(111) are obtained. Samples grown in the Ga-rich regime show better structural and optical quality with reduced density of cubic inclusions and relatively strong photoluminescence emission at 3.45 eV. Threading screw and edge dislocation densities are on the order of 10¹⁰ cm⁻².

Optimization of the initial GaN nucleation behavior on Sc₂O₃ is expected to provide further improvements in the GaN quality, making this engineered oxide buffer approach an attractive route to integrate GaN on Si wafers.

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