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Control growth orientation of semipolar GaN layers grown on 3C-SiC/(001) Si

Duc V. Dinh[a,*], Peter J. Parbrook[a,b]

[a] *Tyndall National Institute, University College Cork, Lee Maltings, Dyke Parade, Cork, Ireland*
[b] *School of Engineering, University College Cork, Cork, Ireland*

Abstract

Heteroepitaxial growth of GaN buffer layers on 3C-SiC/(001) Si substrates (4°-miscut towards [110]) by metalorganic vapour phase epitaxy has been investigated. High-temperature grown $Al_xGa_{1-x}N$/AlN interlayers were employed to control GaN surface orientations. Semipolar GaN layers with $(10\bar{1}1)$, $(20\bar{2}3)$ and $(10\bar{1}2)$ surface orientations were achieved, as confirmed by x-ray diffraction. Due to the substrate miscut, the growth of $(10\bar{1}1)$ layers was twinned along $[1\bar{1}0]_{3C-SiC/Si}$ and $[\bar{1}10]_{3C-SiC/Si}$ while the growth of $(20\bar{2}3)$ and $(10\bar{1}2)$ layers was only along $[110]_{3C-SiC/Si}$. The $(10\bar{1}1)$ layers have rough surface morphology while the $(20\bar{2}3)$ and $(10\bar{1}2)$ layers have mirror-like smooth surface. For all samples with various surface orientations, different photoluminescence peak emission energies were observed at ∼3.45 eV, 3.78 eV and 3.27 eV at 10 K. These emissions are attributed to the near-band edge of hexagonal GaN, basal-plane stacking faults and partial dislocations, respectively. The dominant luminescence intensity of stacking faults indicates their high density in the GaN layers.

Keywords: A3. Metalorganic vapour phase epitaxy, B1. Nitrides, B2. GaN, B2. Semiconducting aluminium compounds

1. Introduction

Group III-nitride semiconductor compounds have attracted much attention for application in optoelectronic devices. Typically, such devices are epitaxially grown along the [0001] direction; however, the distortion of the crystal lattice introduces a large piezoelectric field, which is detrimental to the performance of the devices. This field can be much reduced along semi- and non-polar directions [1].

Compared to sapphire substrates, silicon (Si) substrates are promising for nitride-based optoelectronic devices due to its low-cost large-diameter wafer, and well-characterized electrical and thermal properties. Different semipolar GaN layers and InGaN/GaN light-emitting diodes (LEDs) have already been prepared on different planar Si substrates such as $(10\bar{1}2)$ GaN (with $(10\bar{1}1)$ GaN inclusions) grown on 2-6°-miscut (001) Si [2], $(10\bar{1}6)$ LEDs on (112) Si and $(10\bar{1}5)$ LEDs on (113) Si [3], as well as $(10\bar{1}3)$ GaN [4, 5] and $(10\bar{1}5)$ GaN [5] on (001) Si. To improve material quality, patterned Si substrates have also been used to grow semipolar GaN selectively, e.g., $(10\bar{1}1)$ GaN on patterned (001) Si [6], $(11\bar{2}2)$ GaN on patterned (113) Si [7] and $(20\bar{2}1)$ GaN on patterned (114) Si [8]. However, GaN layers on patterned substrates generally have much rougher surfaces compared to layers grown on planar substrates.

For growth of high-quality GaN films, Si substrates are much less suitable due to a large difference in the in-plane thermal expansion coefficients ($5.6\times10^{-6}\,K^{-1}$ for GaN and $2.6\times10^{-6}\,K^{-1}$ for Si) and a large lattice mismatch (e.g., ∼17% between (0001) *c*-plane GaN and (111) Si). This large thermal expansion mismatch generally leads to cracking in GaN layers during cooling from the growth temperature to room temperature. To achieve crack-free GaN on Si, prior to GaN epitaxy, AlN [2, 6, 7] and AlGaN/AlN interlayers [3] have

*Corresponding author: duc.vn.dinh@gmail.com
Current address: Institute of Materials and Systems for Sustainability, Nagoya University, Nagoya 464-8601, Japan

been employed. Cubic (3C)-SiC has also been used as an intermediate layer for (0001) GaN epitaxy on (111) Si to reduce the lattice and thermal mismatches [9, 10]. Semipolar $(10\bar{1}2)$ GaN layers with 1-μm thickness have been produced on 8°-miscut 3C-SiC/(001) Si with an AlN interlayer by metal-organic vapour phase epitaxy (MOVPE) [11]. However, they are cracked and have a non-uniform surface morphology. Growth of $(20\bar{2}3)$ GaN layer has been attempted on 3C-SiC/(001) Si using hydride-chloride vapour-phase epitaxy [12]. However, the layer consists of oriented grains with size of several tens of microns. Recently, crack-free mirror-like 1.5-μm-thick $(20\bar{2}3)$ GaN layers have been successfully grown on 3C-SiC/(001) Si substrates by MOVPE [13].

In this paper, we report on MOVPE-growth and characterization of GaN layers on 3C-SiC templates, which were prepared on 4°-miscut (001) Si substrates. AlGaN/AlN interlayers were optimized to achieve semipolar $(10\bar{1}1)$, $(10\bar{1}2)$ and $(20\bar{2}3)$ surface oriented GaN layers.

2. Experimental

10-μm-thick 3C-SiC templates grown on 4-inch n-type (001) Si wafers (4°-miscut towards [110]) using low-pressure hot-wall chemical vapour deposition were produced by Anvil Semiconductors [14, 15]. 100-μm-width polycrystalline SiC grid patterns were used to divide the wafers into square coupons ($2.5 \times 2.5\,\mathrm{mm}^2$ size) for stress relief. The full-width at half maximum (FWHM) value of the (002) 3C-SiC symmetric X-ray rocking curve (XRC) of the templates is about 500 arcsec.

Growth of GaN was performed on 3C-SiC/(001) Si substrates in an Aixtron 3×2-inch close-coupled showerhead MOVPE reactor. Ammonia, trimethylaluminium and trimethylgallium were used as precursors. Under a reactor pressure of 50 mbar, the substrates were heated up and thermally cleaned for 2 minutes at ∼1100°C in H_2 ambient. Afterwards, an approximately 10-nm-thick AlN layer was grown on the substrates at 1100°C. The reactor pressure was increased to 200 mbar during cooling to 950°C corresponding to a growth temperature of AlGaN interlayers as measured by a Laytec *in-situ* pyrometer. Approximately 10-nm-thick $Al_xGa_{1-x}N$ interlayers ($x_{AlN} \sim 0$ - 0.6) were grown on the AlN/3C-SiC/Si samples followed by a Si-doped n-type GaN overgrowth. Growth parameters of $Al_xGa_{1-x}N$ are reported elsewhere [16].

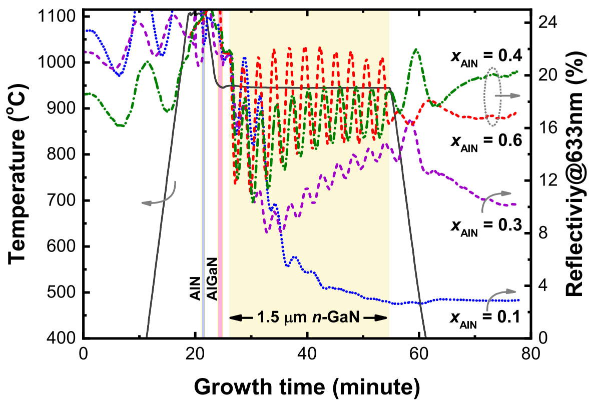

Figure 1: *In-situ* transients of the 633 nm reflectance measured during growth of n-type GaN layers grown on 3C-SiC/(001) Si substrates with different $Al_xGa_{1-x}N$ interlayers.

In-situ analysis was performed using a triple wavelength Laytec EpiTT system giving reflectometry at 405 nm, 633 nm and 950 nm. The crystal orientation and properties of the GaN samples were characterized using a PANalytical X'pert triple-axis high-resolution X-ray diffraction (XRD) system with a $CuK_{\alpha 1}$ source. The surface morphology of the samples was investigated by atomic force microscopy (AFM) in tapping mode (Veeco multimode V) and by Nomarski differential interference contrast microscopy (Olympus XC30). Temperature-dependent photoluminescence (TD-PL) measurements of the samples were performed by a Horiba iHR320 spectrometer using a continuous-wave 244-nm Ar^+ laser as excitation source.

3. Results and Discussion

Fig. 1 shows *in-situ* transients of the 633 nm reflectance recorded during growth of n-type GaN layers grown on the 3C-SiC/(001) Si substrates with different $Al_xGa_{1-x}N$/AlN interlayers. For all samples, damping reflectance has been observed that is attributed to a transition from three-dimensional to two-dimensional growth. This damping has been found to decrease with increasing AlN mole fraction of the $Al_xGa_{1-x}N$ interlayers, suggesting smoother surface morphology. By fitting the Fabry-Pérot oscillations (e.g., for layers grown with $x_{AlN} \geq 0.4$), the thickness of the layers has been estimated to be (1500 ± 100) nm.

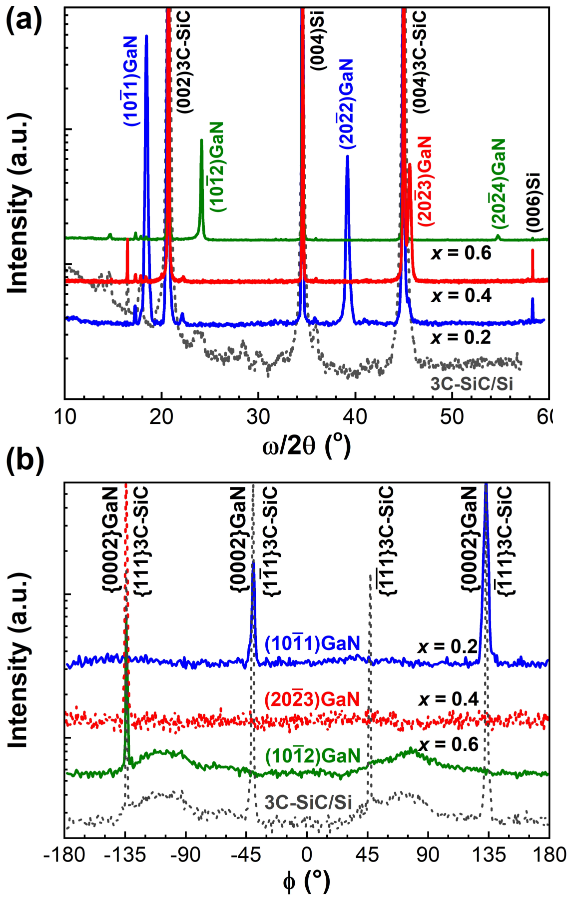

Figure 2: (a) Symmetric $\omega/2\theta$ XRD scans of n-type GaN layers grown on 3C-SiC/(001) Si substrate with different $Al_xGa_{1-x}N$ interlayers ($x_{AlN} = 0.2$, 0.4 and 0.6). (b) XRD off-axis ϕ-scans performed in skew-symmetry with settings for the {0002} reflection of (10$\bar{1}$1) GaN ($x_{AlN} = 0.2$), (20$\bar{2}$3) GaN ($x_{AlN} = 0.4$) and (10$\bar{1}$2) GaN layers ($x_{AlN} = 0.6$), as well as for the {111} reflection of the substrate.

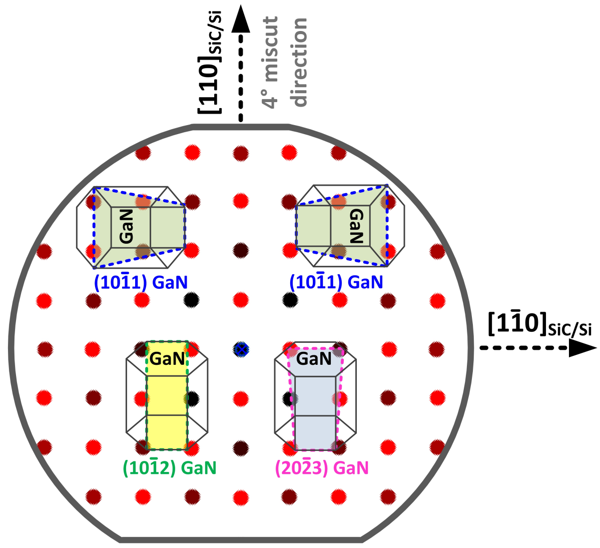

Figure 3: A schematic illustration of three different semipolar (10$\bar{1}$1), (10$\bar{1}$2) and (20$\bar{2}$3) surface oriented GaN possibly grown separately on n-type 3C-SiC/(001) Si (with 4°-miscut towards $[110]_{3C-SiC/Si}$) with three different AlN mole fraction ranges of $x_{AlN} = 0$ - 0.3, 0.4 - 0.5 and 0.5 - 0.6 of $Al_xGa_{1-x}N$ interlayers, respectively.

Fig. 2(a) shows symmetric XRD $\omega/2\theta$ scans performed with an open detector of the GaN layers grown on 3C-SiC/(001) Si substrates with different $Al_xGa_{1-x}N$ interlayers (e.g., $x_{AlN} = 0.2$, 0.4 and 0.6). An XRD scan of the substrates is also plotted for comparison that shows clearly the 3C-SiC ((002) and (004)) and Si ((004) and (006)) reflections, as well as several reflections from SiC grids (e.g., at about 16 and 36°). For the GaN layers grown with $x_{AlN} = 0.2$, 0.4 and 0.6 of the interlayers, different dominant reflections are found at about 18.4°, 45.6° and 24.1°, corresponding to the (10$\bar{1}$1), (20$\bar{2}$3) and (10$\bar{1}$2) reflections of GaN, respectively. It has been found that (10$\bar{1}$1), (20$\bar{2}$3) and (10$\bar{1}$2) layers can be grown by the use of three AlN mole fraction ranges of $x_{AlN} = 0$ - 0.3, 0.4 - 0.5 and 0.5 - 0.6 of the interlayers, respectively. The XRC FWHM value of all GaN layers measured with an open detector without any receiving slit is about 1.0°, indicating their low material quality. This might be due to a propagation of stacking faults from 3C-SiC into the GaN layers [10, 11].

The epitaxial in-plane relationship of the GaN layers with respect to the 3C-SiC/Si substrates was determined from XRD off-axis ϕ-scan. The skew-symmetric {111} reflection of (001) 3C-SiC was measured with a tilt angle ($\psi = 54.7°$), which indicates $[110]_{3C-SiC}$ and $[1\bar{1}0]_{3C-SiC}$. To indicate $[0001]_{GaN}$ of the (10$\bar{1}$1), (10$\bar{1}$2) and (20$\bar{2}$3) layers, the skew-symmetric {0002} reflection was measured with $\psi = 62.0°$, 43.2° and 54.4°, respectively. As shown in Fig. 2(b), for the (10$\bar{1}$2) and (20$\bar{2}$3) layers, only one skew-symmetric $\{0002\}_{GaN}$ reflection was observed indicating that $[0001]_{GaN}$ is pointing along $[110]_{3C-SiC}$. In contrast, for the (10$\bar{1}$1) layers, two {0002} reflections have been observed that rotate by ±90° with respect to the {0002} reflection of the (10$\bar{1}$2) and (20$\bar{2}$3) layers. This indicates a crystalline twinning of the (10$\bar{1}$1) layers, i.e., $[0001]_{GaN}$ is pointing along both $[1\bar{1}0]_{3C-SiC}$ and $[\bar{1}10]_{3C-SiC}$. Fig. 3 shows a schematic illustration of three different (10$\bar{1}$1),

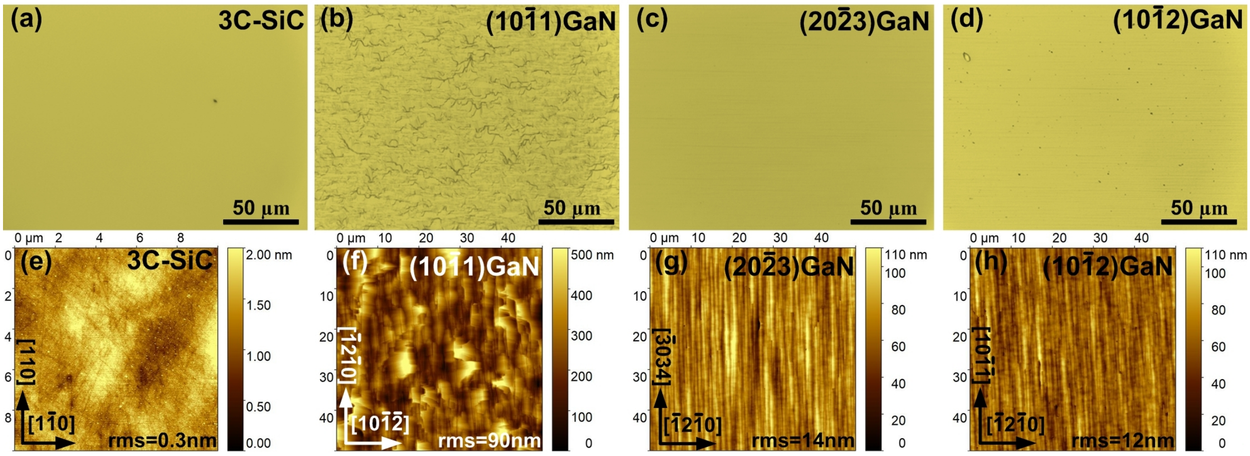

Figure 4: (Top row) Nomarski images of (a) the 3C-SiC/(001) Si substrates, (b) 1.5-μm-thick n-type $(10\bar{1}1)$, (c) $(20\bar{2}3)$ and (d) $(10\bar{1}2)$ GaN layers. (Bottom row) A $20 \times 20\,\mu m^2$ AFM image of the substrates (e) and $50 \times 50\,\mu m^2$ images of the layers grown on the substrates with (f) $(10\bar{1}1)$, (g) $(20\bar{2}3)$ and (h) $(10\bar{1}2)$ GaN surface orientations. Root-mean square (rms) roughness values are shown for comparison.

$(10\bar{1}2)$ and $(20\bar{2}3)$ GaN possibly grown separately on the substrates with three different AlN mole fraction ranges of $x_{AlN} = 0$ - 0.3, 0.4 - 0.5 and 0.5 - 0.6 of $Al_xGa_{1-x}N$ interlayers, respectively.

For $(10\bar{1}1)$, $(10\bar{1}2)$ and $(20\bar{2}3)$ GaN on (001) 3C-SiC, the lattice mismatch of a-lattice between AlN (GaN) and 3C-SiC is estimated to be about 0.9% (3.4%); the lattice mismatch between c'-lattice of AlN (GaN) and 3C-SiC is estimated to be about -4.1% (-1.2%), -15.4% (-10.2%) and 19.0% (22.9%), respectively. Due to the smallest lattice mismatch, $(10\bar{1}1)$ GaN layers should be always formed on non-miscut substrates and they can be grown equally along four directions of 3C-SiC, i.e., [110], $[1\bar{1}0]$, $[\bar{1}\bar{1}0]$ and $[\bar{1}10]$. However, according to the XRD off-axis ϕ-scan (Fig. 2(b)), the $(10\bar{1}1)$ layers were found to grow only along $[1\bar{1}0]$ and $[\bar{1}10]$. This is plausible due to the 4°-miscut towards $[110]_{3C-SiC/Si}$ of the substrates. This miscut also leads to the $(10\bar{1}2)$ and $(20\bar{2}3)$ layers growing only along [110]. Similar findings have previously been observed for $(10\bar{1}2)$ GaN on 2-6°-miscut (001) Si [2] and on 8°-miscut 3C-SiC/(001) Si [11].

Fig. 4 shows the surface morphology of 3C-SiC/(001) Si substrates and the n-type GaN layers with $(10\bar{1}1)$, $(20\bar{2}3)$ and $(10\bar{1}2)$ surface orientations. All layers show a crack-free surface. The root-mean square (rms) roughness of the substrates is about 0.3 nm estimated from a $20 \times 20\,\mu m^2$ scan area. The $(10\bar{1}1)$ layers have rough surface morphology with an rms value of ~90 nm ($50 \times 50\,\mu m^2$). In contrast to the $(10\bar{1}1)$ layers, layers with the $(20\bar{2}3)$ and $(10\bar{1}2)$ surface orientations exhibit smooth morphology with similar rms values of ~14 nm ($50 \times 50\,\mu m^2$). These latter layers have an undulated morphology along $[\bar{1}2\bar{1}0]_{GaN}$ due to the anisotropic diffusion of the group-III atoms on these surfaces. Photographs of the $(10\bar{1}1)$ and $(20\bar{2}3)$ GaN layers grown on 4-inch 3C-SiC/(001) Si wafers are shown in Fig. 5, showing a matt surface of the $(10\bar{1}1)$ wafer and a mirror-like surface of the $(20\bar{2}3)$ wafer (and $(10\bar{1}2)$ wafer - not shown). Strong wafer bowing during GaN growth caused the black visible on the $(20\bar{2}3)$ wafer.

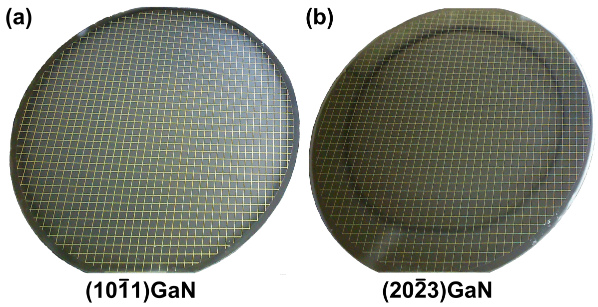

Figure 5: Photographs of the n-type $(10\bar{1}1)$ and $(20\bar{2}3)$ GaN layers grown on 4-inch n-type 3C-SiC/(001) Si wafers.

TD-PL measurements (10 - 300 K) were carried out to investigate the optical properties of the GaN layers with different surface orientations. All layers have typical TD-PL spectra as shown in Fig. 6. Different peak emission energies are observed at

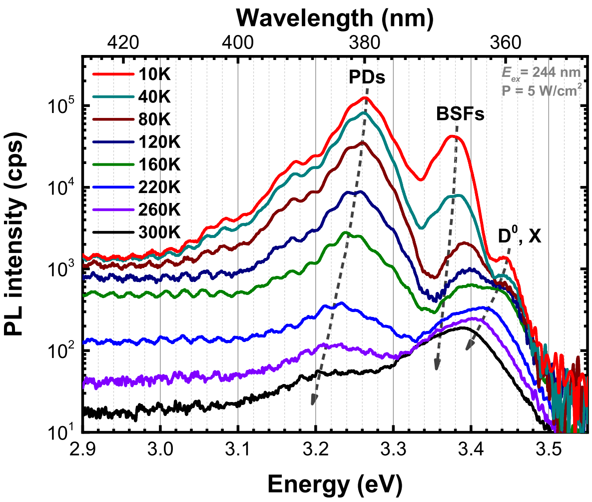

Figure 6: TD-PL spectra of a $(10\bar{1}2)$ GaN layer grown on 3C-SiC/(001) Si substrates.

~3.45 eV, 3.38 eV and 3.27 eV at 10 K. They are attributed to the near-band edge of hexagonal GaN (D^o, X), I_1-type basal-plane stacking faults (BSFs) and partial dislocations (PDs) terminating the BSFs, respectively [17]. The dominant BSF and PD emission intensities indicate their high densities in the layers (i.e., in the range of 10^6 cm^{-1}).

4. Conclusions

Heteroepitaxial MOVPE-growth of crack-free 1.5-μm-thick n-type GaN buffer layers on $Al_xGa_{1-x}N$/AlN interlayers grown on n-type 3C-SiC/(001) Si substrates (4°-miscut towards [110]) has been investigated. By adjusting the AlN mole fraction of $Al_xGa_{1-x}N$/AlN interlayers, GaN layers with three different surface orientations have been achieved including $(10\bar{1}1)$, $(20\bar{2}3)$ and $(10\bar{1}2)$. The miscut has been found to cause twins in the $(10\bar{1}1)$ layers along $[1\bar{1}0]_{3C-SiC/Si}$ and $[\bar{1}10]_{3C-SiC/Si}$, while the growth of $(20\bar{2}3)$ and $(10\bar{1}2)$ layers was only along the miscut direction. The $(10\bar{1}1)$ layers have rough surface morphology, while the $(20\bar{2}3)$ and $(10\bar{1}2)$ layers have mirror-like smooth surface. For all layers, low-temperature PL measurements show dominant stacking fault and dislocation emission intensities indicating their high densities in the layers. Thus, GaN growth conditions need to be further optimized to improve crystallinity.

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