High-temperature molecular beam epitaxial growth of AlGaN/GaN on GaN templates with reduced interface impurity levels

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(Received 27 October 2009; accepted 7 December 2009; published online 25 February 2010)

We present combined in situ thermal cleaning and intentional doping strategies near the substrate regrowth interface to produce high-quality AlGaN/GaN high electron mobility transistors on semi-insulating (0001) GaN templates with low interfacial impurity concentrations and low buffer leakage. By exposing the GaN templates to an optimized thermal dissociation step in the plasma-assisted molecular beam epitaxy environment, oxygen, carbon, and, to lesser extent, Si impurities were effectively removed from the regrowth interface under preservation of good interface quality. Residual Si was further compensated by C-doped GaN via CBr4 to yield highly resistive GaN buffer layers. Improved N-rich growth conditions at high growth temperatures were then utilized for subsequent growth of the AlGaN/GaN device structure, yielding smooth surface morphologies and low residual oxygen concentration with large insensitivity to the (Al+Ga)N flux ratio. Room temperature electron mobilities of the two-dimensional electron gas at the AlGaN/GaN interface exceeded >1750 cm2/V·s and the dc drain current reached ∼1.1 A/mm at a +1 V bias, demonstrating the effectiveness of the applied methods. © 2010 American Institute of Physics. [doi:10.1063/1.3285309]

I. INTRODUCTION

AlGaN/GaN high electron mobility transistors (HEMTs) have recently gained large attention for high-power and high-frequency electronic devices due to their significant merits associated with the large breakdown field, the high saturation velocity, and the high sheet charge density of the polarization-induced two-dimensional electron gas (2DEG). To fabricate nitride-based HEMT materials, molecular beam epitaxy (MBE) has become an established growth technique for state-of-the-art devices1–4 as it offers many benefits, such as sharp interface and doping control, low background impurity incorporation, and powerful in situ diagnostics for precise growth control.

Despite these promises, direct growth by conventional plasma-assisted (PA)-MBE, typically performed under metal-rich conditions at fairly low temperatures, was often hampered by the huge requirements to provide low threading dislocation densities (TDDs) in the GaN buffer for low device leakage along with smooth, homogeneous AlGaN/GaN heterostructures. Only through complex multistep buffer layers for effective threading dislocation (TD) reduction during heteroepitaxial nucleation on foreign substrates [i.e., sapphire5 and SiC (Ref. 6)] and optimized growth conditions around the narrow boundary for Ga droplet formation were successes achieved with atomically smooth metal and surface pit-free heterointerfaces.7,8

Most recently, growth in nitrogen-rich environment by PAMBE (Ref. 9) and ammonia-based MBE (Refs. 10 and 11) delivered significant advances. In particular, these were wider growth windows and potentially higher growth temperatures (£760 °C) with suppressed thermal decomposition9–13 and lower impurity incorporation,14 resulting in TDDs reduced to <5 × 109 cm−2.15,16 the highest reported room temperature (RT) bulk electron mobilities (μ > 1150 cm2/V·s)5 in MBE-grown GaN and exceptional AlGaN/GaN HEMT power performance on SiC and Si.14,17

In addition, under N-rich growth conditions reduced leakage currents were reported due to the lack of excess Ga decoration along TDs.18

Ultimately, to increase the intrinsic properties of the 2DEG in AlGaN/GaN structures and to reduce leakage paths, the number of charged TDs propagating through the device needs to be further minimized via higher-quality substrates. Both the use of semi-insulating (SI) or freestanding GaN templates grown by metal organic vapor phase epitaxy or hydride vapor phase epitaxy on sapphire resulted recently in RT 2DEG mobilities >1900 cm2/V·s and record μ ~ 167 000 cm2/V·s at 0.3 K in MBE-regrown AlGaN/GaN heterostructures due to the overall lower TDDs (∼107–8 × 108 cm−2).19–21

Although with further development of SI and self-supporting low-TDD GaN templates, parasitic conduction at the homoepitaxial regrowth interface (RI) presents an independently serious problem. This arises from the typical interface contamination (mainly O, Si, and C impurities) adsorbed during air exposure in standard substrate loading procedures or at the beginning of regrowth, requiring routes to eliminate the n-type conductive channel close to the MBE-GaN-template interface. Wet chemical etching in HF, HCl, KOH, and sulfuric acids, together with thermal desorption via in situ vacuum annealing at elevated temperatures...
yielded only limited reduction of either O or C impurities.\textsuperscript{22–23} Instead, annealing studies in NH\textsubscript{3} ambient at 700–800 °C resulted sometimes in sufficiently clean and stoichiometric (0001) GaN surfaces, although not performed in a closed MBE growth environment.\textsuperscript{24} Most of these in situ annealing methods were additionally constricted by the very low thermal dissociation temperature of the (0001) GaN surface, i.e., \(\sim750 ~°C \) in vacuum.\textsuperscript{12,13,24}

More effective compensation of the interfacial impurities yielding highly insulating GaN buffer layers in the regrown MBE-AlGaN/GaN structures was demonstrated by the use of different acceptors, such as beryllium\textsuperscript{19} and carbon,\textsuperscript{26,27} or through graded iron concentration profiles in the underlying Si GaN templates.\textsuperscript{17} Nevertheless, compensation-doped GaN buffer layers need to be well balanced in thickness and doping concentration for sufficient isolation and to minimize dispersion effects in the 2DEG at the AlGaN/GaN heterointerface.

In the present study, we merged many of the critical aspects mentioned above to fabricate high-quality AlGaN/GaN heterostructures on low-dislocation-density (0001) GaN templates with improved RI properties and low buffer leakage. Essentially, we developed a sophisticated thermal dissociation procedure prior to GaN buffer nucleation, eliminating both oxygen and carbon impurities at the RI directly in the MBE growth environment and without the use of wet chemical etchants. The common propensity for residual Si remaining at the RI was solved by additional compensation doping via highly insulating C-doped GaN buffer layers. Growth of the AlGaN/GaN heterostructures on top demonstrated further the significant merits of the N-rich growth conditions employed in the thermal decomposition regime, with RT 2DEG mobilities \(>1750 \text{ cm}^2/\text{V} \cdot \text{s} \) and growth surfaces free from detrimental excess Ga.

## II. EXPERIMENTS

As substrates we used semi-insulating (Fe-doped) \(\sim3.5-\mu\text{m-thick} \) (0001) GaN templates grown on c-plane sapphire by MOCVD (Lumilog). The TDD and resistance were specified as \(\sim5 \times 10^{10} \text{ cm}^2\) and \(\sim10 \text{ M} \Omega \) at RT, respectively. For sample preparation, the 2 in. GaN substrates’ backsides were metalized with \(\sim500-\text{nm-thick Ti} \) to enable radiative heating, then cleaved into 0.5–2 in. pieces, and subsequently degreased in acetone, methanol, and isopropanol. For reference, two samples were additionally etched in buffered hydrofluoric and hydrochloric acids (HF and HCl) to investigate the effect of potentially reduced substrate surface contamination. After blow drying under N\textsubscript{2} ambient, the samples were immediately loaded into the loadlock of the MBE chamber and were outgassed for 1 h at 400 °C prior to growth.

All further experiments were carried out in situ in a Gen-II MBE system equipped with standard effusion cells for Ga and Al and a Veeco Unibulb radio frequency plasma source for supplying active nitrogen. The N\textsubscript{2} gas was specified with 7N purity (99.99999%) and cleaned via two inert gas purifiers at the gas flow inlet prior to entering the plasma source. The substrate temperature was measured in situ via an optical pyrometer. For intentional carbon doping, we used a commercially available carbon tetrabromide (CBr\textsubscript{4}) sublimation system, where the variable foreline pressure, regulated through automated valve control, was employed to control the CBr\textsubscript{4} flow. Except for the CBr\textsubscript{4} flow (in foreline pressure units of mTorr), we gave all other molecular fluxes in (0001) GaN growth rate units (nm/min), as calibrated from thickness measurements of Ga- and N-limited GaN films at temperatures of negligible thermal decomposition.\textsuperscript{28} For conversion, we note that in wurzite GaN \(c/2 = 0.259 \text{ nm} \) or \(1.14 \times 10^{15} \text{ GaN/cm}^2\) areal density refer to 1 ML. Moreover, two specific N fluxes were used, i.e., \(\sim3.5 ~\text{ nm/min} \) [250 W/0.3 SCCM (SCCM denotes standard cubic centimeter per minute)] during the initial thermal dissociation procedure and subsequent C-doped GaN buffer growth, and \(\sim5 ~\text{ nm/min} \) (300 W/0.4 SCCM) for the undoped N-rich/high-T grown AlGaN/GaN heterostructure on top. Since the Ga/N flux ratio for the AlGaN/GaN structure was at \(\sim0.75–0.9\),\textsuperscript{29} the growth rate during the entire structure was nearly identical. A schematic of the typical sample structure and the thermal dissociation step are illustrated in Fig. 1.

![FIG. 1. (Color online) (a) Schematic of typical MBE–grown AlGaN/GaN heterostructure on MOCVD-GaN template (as referred to also in Figs. 7 and 8), utilizing C-doped GaN buffer layer. N-rich growth conditions in the GaN thermal decomposition regime, and (b) an optimized thermal cleaning step at the RI prior to layer growth. This step consists of alternating cycles of 1 min (1’) GaN growth and 2 min (2’) thermal decomposition under vacuum at \(T=780 °C\). Labels “g.r.” and “d.r.” refer to the growth and decomposition rates, respectively. The net amount of evaporated GaN during one such thermal cleaning cycle corresponds to 2.5 nm.](image)

Reflection high energy electron diffraction (RHEED) was employed to monitor the surface properties throughout the thermal dissociation procedure at the RI and the film growth. The effects of these individual growth steps on the concentration of interfacial impurities (O, C, and Si) and intentional carbon dopants were further analyzed by secondary ion mass spectroscopy (SIMS). The surface morphologies of the thermally treated GaN templates and the final AlGaN/GaN heterostructure were characterized by atomic force microscopy (AFM). The quality of the AlGaN/GaN heterointerfaces and the Al content in the AlGaN top layer was further analyzed using high-resolution x-ray diffraction (HRXRD). Finally, test structures were fabricated using contact photolithography, including van der Pauw (vdP) patterns for measurements of the 2DEG mobilities and test patterns for the evaluation of isolation buffer leakage currents and maximum drain currents.

## III. RESULTS

### A. Thermal cleaning of GaN template

The in situ thermal dissociation procedure applied to the (0001) GaN template required a careful counterbalance be-
between (i) effective thermal desorption of surface impurities and (ii) preservation of smooth surface morphology. In general, we found that keeping the GaN template in vacuum at substrate temperatures below the onset of thermal dissociation, i.e., \( \sim 750 \, ^\circ\text{C} \),\(^{12,13} \) for duration of several minutes to up to 1 h, resulted in no discernible reduction of the O, C, and Si-related surface impurities. This is evident in the SIMS profiles of Fig. 2(a), showing increased levels of O \( \left( 5.5 \times 10^{19} \, \text{cm}^{-3} \right) \), C \( \left( 5.0 \times 10^{19} \, \text{cm}^{-3} \right) \), and Si \( \left( 2.0 \times 10^{19} \, \text{cm}^{-3} \right) \) at the RI between the GaN template and the top MBE-GaN layer grown at 700 \, ^\circ\text{C}. These large O, C, and Si concentrations remained also unaffected by etching the GaN template either in buffered HF or in HCl acids prior to loading into the MBE chamber.

On the other hand, elevated substrate temperatures of \( T > 760 \, ^\circ\text{C} \) yielded a characteristic layer-by-layer thermal dissociation behavior in vacuum evidenced by RHEED intensity oscillations recorded along the \([1120]\) azimuth with periods corresponding to the evaporation of individual MLs (not shown here). According to our previous investigation, the layer-by-layer thermal dissociation rate obeyed an exponential behavior with substrate temperature.\(^{30} \) However, the layer-by-layer-like evaporation ceased readily with the appearance of slightly faceted surfaces after the dissociation of several MLs, in particular for \( T > 800 \, ^\circ\text{C} \). The critical time for the onset of facet formation—as observed by RHEED—

was approximately 1.5 min at 800 \, ^\circ\text{C} and 2.5 min at 780 \, ^\circ\text{C}, equivalent to the evaporation of \( \sim 15–20 \, \text{ML} \) of GaN based on our in situ measurements.\(^{30} \)

Utilizing such single-step thermal dissociation procedure below the limits of facet formation did not reproducibly desorb the surface impurities to below the detection limit of SIMS. To access lower impurity concentration at the GaN template surface, we developed an optimized thermal cleaning procedure that consisted of alternating vacuum thermal dissociation and GaN regrowth cycles at constant 780 \, ^\circ\text{C}. As depicted in Fig. 1(b), after raising the substrate temperature to 780 \, ^\circ\text{C}, the cycles were initiated with 1 min of GaN growth under slightly Ga-rich conditions, followed by 2 min of thermal dissociation in vacuum, with closed Ga and N shutters. This sequence was repeated several times, where the shorter GaN regrowth step essentially helped to preserve the surface from increased facet formation. The net growth rate of the GaN regrowth step was 1.5 nm/min (i.e., 3.5 nm/min supplied N flux minus 2 nm/min thermal dissociation rate at 780 \, ^\circ\text{C}), while the thermal dissociation step delivered a constant GaN desorption rate of 2 nm/min. Thus, one cycle resulted in the net GaN evaporation of 2.5 nm, i.e., \( \sim 10 \, \text{ML} \).

The SIMS profile in Fig. 2(b) demonstrates the effective reduction of both oxygen and carbon impurities at the GaN template interface by using ten cycles of the proposed procedure (i.e., 25 nm net amount of evaporated GaN). Reproducible reduction by three orders of magnitude of both O and C levels to the detection limit of SIMS \( \left( [\text{O,C}] \sim 10^{16} \, \text{cm}^{-3} \right) \) was achieved for cycle periods of 5, 10, and 20, corresponding to \( \sim 12.5–50 \, \text{nm} \) thickness etched thermally from the GaN template surface. Note that the Si impurity level at the interface was barely affected by this procedure. This was not surprising due to the extremely low vapor pressure of Si at these temperatures, and strategies to overcome Si interface contamination will be discussed below.

The surface morphology during the repeated regrowth/thermal dissociation cycles showed no significant roughening or faceting effects. This was confirmed by the continuously streaky, i.e., two-dimensional (2D), RHEED patterns and ex situ by AFM scans of the treated GaN template surfaces after immediate cool-down (Fig. 3). Surprisingly, a comparison with the untreated GaN template surface evidenced no increase in surface root-mean-square (rms) roughness even for increased cycle periods, with typical rms val-
values of less than 0.5 nm over a $3 \times 3$ $\mu m^2$ area. However, the step-flow growth features present on the untreated GaN template surface were diminished, and morphologies with 2D islandlike growth features formed instead. These are clearly obvious in Fig. 3(c), showing a large density of 1 ML (i.e., $\sim 0.3$ nm) high islands of arbitrary shape, which is consistent with the preferential 2D layer-by-layer growth and dissociation mode observed under high-$T$ MBE growth.

Furthermore, the treated GaN template surfaces exhibited several randomly distributed shallow surface pits, with depths of $\sim 1$–2 nm and densities of $4 \times 10^8$ cm$^{-2}$ [Fig. 3(b)] and $6 \times 10^8$ cm$^{-2}$ [Fig. 3(c)]. These surface pit densities are identical to the total TDD of the GaN template, confirming the common association of shallow surface pits in MBE-grown GaN with the surface termination of TDDs.$^{28,31}$ We thus proved that the thermal cleaning procedure preferentially etches along these surface defects, unveiling the underlying TD network, similar to wet chemical or photoelectrochemical techniques.$^{32}$ Given the similar surface pit widths and depths in Figs. 3(b) and 3(c), no correlation between the total amount of GaN evaporated and surface pit-size effects could be identified. This provides, moreover, an effective thermal etching route of (0001) GaN layers without significant changes in surface morphology and applicable even toward larger etch depths.

**B. Compensation doping of residual Si**

Although the dominant oxygen and carbon impurities were eliminated from the RI by this procedure, residual Si with concentrations of $10^{18}$–$2 \times 10^{19}$ cm$^{-3}$ were still present. These [Si] are expected to yield yet increased buffer leakage,$^{21,32}$ as shown further below, limiting the isolation of the 2DEG at the AlGaN/GaN interface from the parasitic capacitances at the MBE-GaN/GaN-template RI. To achieve superior electrical isolation for better rf performance of the AlGaN/GaN devices, we investigated the use of C-doped GaN (GaN:C) buffer layers, which were also recently employed for AlGaN/GaN structures on sapphire,$^{33}$ SiC,$^{26,34}$ and Si (111) (Ref. 27) substrates. As illustrated in Fig. 1(a), the doped GaN:C layer was 400 nm thick and directly grown on the GaN template after the completion of the thermal dissociation cycles. Variable CBr$_4$ foreline pressures were used to identify the carbon incorporation efficiency for the GaN buffer layers. Also, the growth conditions for the C-doped GaN layers were varied between the two sophisticated GaN growth regimes, i.e., Ga-rich, low-$T$ (Ga/N=1.7, $T=680$ °C) and N-rich, high-$T$ (Ga/N=0.85, $T=790$ °C). As indicated earlier, the GaN:C buffer layer growth was followed by undoped $\sim$30-nm-thick AlGaN/1-μm-thick GaN device layer grown under constant N-rich, high-$T$ conditions (Ga/N=0.85, $T=790$ °C).

The SIMS data in Fig. 4 show representative C-doping profiles in relation to the interfacial [Si] for these two buffer layer growth regimes. Most importantly, carbon incorporation was at least two orders of magnitude higher under Ga-rich/low-$T$ conditions compared to N-rich/high-$T$ conditions, although the effective buffer layer growth rates were similar. Also, under the N-rich/high-$T$ conditions, the carbon concentration remained largely constant over a large range of CBr$_4$ foreline pressures. We therefore assume that carbon incorporation is much more sensitive to growth temperature than the Ga/N flux ratio, as noted also in a recent study based on lower GaN growth temperatures.$^{35}$

The buffer leakage was investigated by measurements of the drain-source $I$-$V$ curves from isolation patterned AlGaN/GaN HEMT structures with the 2DEG etched away at the gate region. Details on our standard AlGaN/GaN HEMT device dimensions and processing steps are reported elsewhere.$^{4,26}$ Figure 5 shows the drain-source $I$-$V$ curves from isolation patterned samples grown (a) without the thermally etched RI and without the C-doped GaN buffer layer (standard sample), (b) with the RI thermally etched by 25 nm (i.e., ten thermal dissociation cycles), but without the

![FIG. 5](https://example.com/fig5.png) Drain-source buffer leakage currents from isolation patterned samples dependent on different MBE-GaN/MOCVD-GaN-template conditions (i.e., with and without thermally cleaned RI and with C-doped GaN buffer layer—the C doping level in this particular sample was $\sim 2 \times 10^{19}$ cm$^{-3}$ as measured by SIMS). The dashed line at 20 V bias is a guide to the eye for comparison between samples.
C-doped GaN buffer layer, and (c) with both the thermally etched RI (again 25 nm) and the C-doped GaN buffer layer. The carbon concentration [C] in this particular GaN:C buffer layer was \( \sim 2 \times 10^{19} \) cm\(^{-3}\), as measured by SIMS, although buffer layers investigated with slightly less or higher [C] gave similar results.

We observed a substantial decrease in buffer leakage current over several orders of magnitude among these three different samples. While the standard sample showed relatively high buffer leakage currents of \( \sim 100 \) mA/mm at 20 V, these leakage currents were sequentially reduced to \( \sim 1 \) mA/mm at 20 V for the sample with its RI thermally etched and even more notably to \( \sim 1 \) \( \mu \)A/mm at 20 V when a C-doped GaN buffer layer was additionally introduced. In comparison, standard samples with additional treatment of buffered HF or HCl prior to growth (not shown here) gave similarly high buffer leakage currents as in the case of the untreated samples. On the other hand, increasing the number of thermal dissociation cycles by a factor of 2, i.e., increasing the etching depth at the GaN template to \( \sim 50 \) nm, could not yield further improvement in buffer leakage currents. This suggests that the residual [Si] at the RI is the main cause of buffer leakage in samples without C-doped GaN buffer layers. Despite the obvious merits of the thermal etching procedure, it is important to note that sufficiently low buffer leakage currents for device operation (\( \leq 1 \) mA) require additional compensation of the residual Si by using C-doped GaN buffer layers.

C. High-temperature AlGaN/GaN growth on GaN template

Despite the recently highlighted N-rich/high-\( T \) growth regime with optimized surface morphologies and electron mobilities for homoepitaxial (0001) GaN layers on GaN templates,\(^9\) the effect of these growth conditions on the growth of AlGaN layers has remained unexplored. Representative AFM images in Fig. 6 illustrate the influence of different (Al+Ga)/N flux ratios [in the range of 0.75 < (Al + Ga)/N < 1.6] on the surface morphology of AlGaN/GaN layers grown at a constant growth temperature \( T=780 \) °C. Note that the varying (Al+Ga)/N flux ratios cover growth conditions from N-rich to Ga-rich growth via adjusting both Al and Ga fluxes such that the resulting \( \sim 30\)-nm-thick AlGaN layers had comparable Al content in the range of 0.34 < \( x \)(Al) < 0.39. Here, the underlying GaN layer was grown under an identical Ga/N flux ratio of 0.75, such that the nucleation of the consecutive AlGaN layers was not obscured by the underlying GaN morphology.

Surprisingly, the AFM images show no discernible differences in morphology among the individual AlGaN samples, yielding atomically flat surfaces with rms roughnesses typically between 0.6 and 0.8 nm (10 \( \times \) 10 \( \mu \)m\(^2\) scan area). Also, all AlGaN surfaces replicate the underlying TDDs, indicated by the shallow surface pit densities of \( \sim 5 \times 10^{8} \) cm\(^{-2}\). This wide growth window for atomically smooth AlGaN layers is in stark contrast to the observed sensitivity of AlGaN surface morphology and surface defect structure to the (Al+Ga)/N flux ratio at lower growth temperatures.\(^36,37\)

Moreover, we investigated the incorporation of unintentional oxygen into both GaN and AlGaN layers grown at high temperatures under both N-rich and Ga-rich conditions. The SIMS data in Fig. 6(c) show the oxygen and Al profiles (as guide to the eye) across an AlGaN/GaN sample structure (see inset) grown under the two distinct high-\( T \) (N-rich versus Ga-rich) growth conditions, relative to low-\( T \) growth conditions. Initiating the growth within this structure by Ga-rich/low-\( T \) conditions and changing these sequentially to Ga-rich/high-\( T \), N-rich/high-\( T \), and N-rich/low-\( T \) conditions, care has been taken to maintain a smooth growth front up to the typically kinetically roughened N-rich/low-\( T \) grown layer. The layer thicknesses for each individual growth condition sequence were 250 nm for GaN and 80 nm for AlGaN, while the Al content of the AlGaN layer was adjusted to \( x \)(Al) \( \sim 0.2 \) to prevent large tensile strain formation and cracking of the AlGaN layers.

A comparison among these AlGaN/GaN layer sequences indicates that the oxygen levels are very similar among the individual AlGaN and GaN layers grown under Ga-rich/low-\( T \), Ga-rich/high-\( T \), and N-rich/high-\( T \) condi-
tions, though oxygen incorporation was slightly higher in AlGaN than in GaN. The N-rich/low-T growth conditions yielded significantly higher oxygen levels (about one order of magnitude higher), which is in agreement with previous reports and will be discussed later.

Due to the independence of the AlGaN surface morphology with the \((\text{Al}+\text{Ga})/\text{N}\) ratio at high growth temperature, we fabricated a simple AlGaN/GaN-on-GaN-template HEMT structure using an \((\text{Al}+\text{Ga})/\text{N}\) flux ratio of 0.9, which yielded an AlGaN thickness of 24 nm and an Al content of 0.38 at the given conditions. Both AlGaN thickness and Al content were calculated based on simulations of the peak separation between GaN and AlGaN peaks in the \(\omega/2\theta \) HRXRD scans under the assumption of a coherently strained AlGaN layer (Fig. 7). Indicative of the high-quality AlGaN/GaN heterostructure, we observed thickness fringes up to at least second order in the HRXRD data. As expected, this was also proven by the atomically smooth surface morphology with rms roughness of less than 1 nm (see inset: AFM image). RT Hall measurements on several vDp patterns fabricated on this sample yielded an average 2DEG Hall mobility of 1760 cm\(^2\)/Vs at a sheet density of \(1.2 \times 10^{13} \text{ cm}^{-2}\) and a sheet resistance of 295 \(\Omega/\square\). We attribute the high mobility observed in this sample to a combination of decently low TDD (mid-10\(^6\) cm\(^2\)/s), good isolation, and low dispersion effects of the 2DEG via reduced impurities at the RI (using the thermal dissociation step) and the 400-nm-thick C-doped GaN buffer layer, as well as the improved N-rich/high-T growth conditions. The high-quality transistor structure was further corroborated by the dc drain current characteristics shown in Fig. 8. The saturation drain current was measured as high as 1.1 A/mm for \(V_G = +1 \text{ V}\), although self-heating effects might be responsible for the slight decrease in drain current at higher voltages. We note that these data compare favorably with the best devices grown on both GaN templates and SiC substrates.

IV. DISCUSSION

This work demonstrates that despite the rapid development of ever lower TDDs of self-supporting GaN templates, parasitic conduction at the RI plays a still dominant role for the intrinsic properties (current leakage paths, 2DEG mobilities, and dc characteristics) of direct-regrown AlGaN/GaN HEMT structures. Since both origin and magnitude of residual free carriers at the near-RI region in these structures seem to affect the buffer leakage characteristics drastically (see Fig. 5), the aim and design of experiments were therefore threefold: (i) to clarify the possible origin and magnitude of residual free carriers, (ii) to reduce the buffer leakage via thermal cleaning of the GaN template and via carbon doping of the GaN buffer, and (iii) to investigate the effect of high-temperature growth conditions on the incorporation of unintentional impurities (specifically oxygen).

Although a number of groups have investigated the first two points, there is yet no standard route of preparing the GaN template and eliminating the parasitic conductive channel at the RI. In the simple thermal (i.e., dry) cleaning procedure of the GaN template introduced in this study, we avoided any wet chemical and/or UV/O\(_3\) treatments, as commonly utilized for GaN surface cleaning prior to Ohmic and Schottky contact deposition. This was motivated mainly by the limited reproducibility of these treatments, which were found to be heavily dependent on the sequence and solution ratios of the applied chemicals (mainly HCl, HF, H\(_2\)SO\(_4\),- and H\(_3\)PO\(_4\)-based solutions) and their frequent traces of additional elements (such as Cl and F) on the surfaces. Furthermore, UV/O\(_3\) treatments were reported to increase the O concentration on the surface despite the effectiveness in removing C. Nevertheless, common to most of these studies were the findings that thermal desorption by high-temperature (\(>700^\circ\text{C}\)) annealing in vacuum, NH\(_3\), N\(_2\), or N\(_2\)/H\(_2\) plasmas after wet chemical and/or UV/O\(_3\) treatments, provided a more effective (although not complete) removal of residual contaminants, while no comments on the effects on surface morphology and electrical properties of devices fabricated were made.

In our thermal cleaning method, large requirements were posed on balancing the effective desorption of surface impurities during the evaporation of GaN and preserving atomically smooth surface morphology. This becomes very impor-
tant as long-period high-temperature thermal cleaning (at temperatures beyond thermal dissociation) is known to introduce large damage to the surface, usually making the GaN material electrically unsuitable.\textsuperscript{41} We noticed that the critical time period for thermal dissociation in vacuum before deterioration of the GaN surface (i.e., \( \sim 2.5 \) min at 780 °C—equivalent to \( \sim 15–20 \) ML of evaporated GaN) was insufficient to thermally desorb all residual O and C to below the detection limit of SIMS. In contrast, the implementation of a short Ga-rich regrowth step or multiple alternating regrowth/dissociation cycles at 780 °C reduced both O and C more effectively. In part, this can be explained by the ready reaction of surface Ga with residual O, forming highly volatile Ga\(_2\)O\(_3\).\textsuperscript{38} We note that applying pure Ga-assisted thermal cleaning without active nitrogen (i.e., “Ga polishing”\textsuperscript{42}) might also be sufficient for the desorption of residual O from the GaN template at 780 °C; however, this simultaneously leads to rough surface morphologies due to the increased thermal dissociation rates under large excess Ga.\textsuperscript{30} It is further important to note that the thermal cleaning procedure was optimized for a fixed N flux of 5 nm/min. With increases in effective N flux, we expect decreases in the thermal dissociation rates such that the thermal cleaning step could shift to much higher temperatures and lead possibly to more enhanced thermal desorption of both O and C.

Obviously, in Fig. 2, the thermal dissociation cycles also exposed surface pits associated with underlying TDs. Since both the surface pit density matched the total TDD of the underlying GaN template and no discernible differences in surface pit sizes were observed for various periods of high-\( T \) dissociation/regrowth cycles, no relation of preferential thermal dissociation on the type of dislocation could be elucidated. This is in contrast to the typically observed pit-size differences and their association with individual types of TDs in MBE-grown GaN layers at low growth temperature.\textsuperscript{31} It is important to note that nearly constant surface pit sizes were also observed in thick GaN epilayers grown by MBE either under N-rich\textsuperscript{9} or Ga-rich conditions at high growth temperatures of \( \sim 780 \) °C.\textsuperscript{45} We therefore attribute the appearance of these surface pits to specific surface kinetics under these high-\( T \) conditions and assume that their sizes might be predefined at the onset of high-\( T \) GaN nucleation or during the initial thermal dissociation step. Nevertheless, we expect that both the duration of the thermal dissociation step and the Ga/N flux ratio during the intermediate regrowth steps are important parameters governing both widening/facetting and filling mechanisms of surface pits, respectively. In the extreme case of prolonged thermal dissociation at high enough temperatures, thermal dissociation along the sidewalls of these surface pits was found to dominate over the 2D layer-by-layer-like dissociation from the planar (0001) GaN surface, resulting in highly faceted GaN surfaces.\textsuperscript{30} During the short intermediate regrowth cycles, however, the Ga-rich conditions seemed to promote a high enough growth rate across these pits preventing rapid surface pitting. The equilibrium between the two counteracting mechanisms could be related to the same Ga-adlayer-mediated surface diffusion enhancement, as recently found for the small pit sizes of (0001) GaN surface morphologies grown by PAMBE under Ga-rich conditions.\textsuperscript{78,31,44}

With residual Si remaining largely unaffected by the thermal cleaning procedure [Fig. 2(b)], the buffer leakage was not reduced to the \( \sim \mu A/\text{mm} \) levels ultimately required for device operation (Fig. 5). As expected, the negligible thermal desorption of Si can be related to its very low vapor pressure in the investigated temperature region. This could be overcome only by significantly larger temperatures (\( >1100 \) °C) in combination with pre-oxidized Si (i.e., SiO\(_3\)) via UV/O\(_3\) or wet chemical etching in HF-based solutions. We have attempted both these methods prior to thermal cleaning; however, the removal of residual Si was unsuccessful due to the limited substrate temperature heating (\( \sim 900 \) °C) of our MBE system.

To compensate residual Si at the RI effectively, carbon was introduced as an intentional dopant to realize semisolating GaN buffer layers.\textsuperscript{26,27,33,34} The marked difference in carbon incorporation and compensation effect within GaN:C buffer layers grown under either Ga-rich/low-\( T \) conditions or N-rich/high-\( T \) conditions can be explained two-fold. First, the incorporation efficiency of carbon via the CBr\(_4\) source was recently found to be strongly temperature dependent and much less dependent on the Ga/N flux ratio.\textsuperscript{35} This is particularly corroborated by the independence of the generally low C incorporation at variable CBr\(_4\) fluences under high-temperature growth conditions, as found in Fig. 4(c). Second, the low compensation effect of carbon under N-rich/high-\( T \) GaN growth conditions is not expected to arise from changes in the amphoteric behavior of carbon in GaN.\textsuperscript{45,46} As GaN layers grown under N-rich/high-\( T \) conditions show similar unintentional \( n \)-type conductivity as those grown under standard Ga-rich/low-\( T \) conditions,\textsuperscript{9} the formation of substitutional C\(_N\) as a shallow acceptor should hold true independent of the given growth conditions.

Another possible effect for low compensation of residual impurities and higher buffer leakage could be related to codoping of unintentional oxygen under N-rich conditions, as evidenced by Green et al.\textsuperscript{35} However, the SIMS profiles of N-rich layers grown under the much higher temperatures in this study did not indicate any measurable increases in oxygen concentration within the GaN:C buffer layers. It is worth noting that the SIMS profiles in Figs. 4(a) and 4(b) show both a distinct low intensity Si peak ([Si] \( \sim (2−3) \times 10^{17} \) cm\(^{-3}\)) at the beginning and end of the GaN:C buffer layer, respectively. In the case of the Ga-rich/low-\( T \) GaN:C buffer layer growth, this can be attributed to residual Si being carried by the Ga-rich GaN:C growth surface, leaving a spike where growth was interrupted and conditions changed to N-rich/high-\( T \) conditions. Thus, residual Si can either diffuse from the RI and be carried on the metal-rich growth surface or be buried by the N-rich growth surface, such that the Si from the RI becomes redistributed between the RI itself and the GaN:C buffer layer. Very similar observations were found for progressive and inhibited Si diffusion at Al-rich versus N-rich AlN surfaces.\textsuperscript{26}

Although the N-rich/high-\( T \) growth conditions have now been widely investigated for GaN epilayers,\textsuperscript{9,12,29,30} their ef-
fect on the growth of AlGaN layers as presented in this study requires further discussion. The apparent independence of the AlGaN surface morphology on the (Al+Ga)/N flux ratio at the high growth temperatures of $\sim 780 - 790 \, ^\circ C$ [at least for AlGaN layers with Al content of less than 40% and 0.75 < (Al+Ga)/N < 1.6] suspects at first glance that the growth kinetics mimic those observed for high-$T$ GaN growth by PAMBE.\cite{9,43} The common surface characteristics given in this wide growth window are consistent with typical 2D layer-by-layer growth features and the lack of stepped terraces and hexagonal spiral hillocks (as under low growth temperatures). It has been argued that these 2D surface morphologies achieved under high growth temperature were mainly due to thermally activated surface diffusion, rather than adlayer mediated diffusion\cite{8} since no surfactant Ga adlayer could be confirmed from in situ RHEED and desorption mass spectrometry analysis.\cite{12} Thus, we suggest that for low-to-intermediate Al-content AlGaN layers, the surface growth kinetics are dominated by the growth temperature, while the effects of varying Al flux are negligible, provided that Al adatoms are fairly mobile at these high growth temperatures.\cite{47}

The similarities in high-$T$ growth kinetics between GaN and AlGaN layers are further corroborated by the incorporation of impurities. Although unintentional oxygen levels were higher in AlGaN relative to GaN layers due to the larger affinity of oxygen in Al-containing layers, the nearly identical oxygen levels within AlGaN layers grown under Ga-rich/low-$T$, Ga-rich/high-$T$ and N-rich/high-$T$ conditions point to the largely independent oxygen incorporation among these regimes (i.e., oxygen sticking coefficient=1 for Al-GaN). This can be explained either by Ga-adlayer mediated growth for Ga-rich conditions\cite{38} or due to substantial thermal desorption at temperatures of thermal dissociation (>750 °C) even for N-rich growth. Hence, oxygen levels were only increased under N-rich/low-$T$ conditions, where both suggested mechanisms were absent.\cite{38}

Considering the similarities of smooth morphology and low oxygen levels in AlGaN layers grown under high-$T$ conditions with those grown under standard Ga-rich/low-$T$ conditions has enabled the fabrication of high mobility 2DEGs and low sheet resistances, even for fairly high Al-content AlGaN/GaN HEMT structures. With the Al content in the AlGaN cap layer varying at x(Al)~0.34–0.38, and respective sheet carrier densities of $>1 \times 10^{13}$ cm$^{-2}$ in the 2DEG, these parameters were rather high for producing optimized 2DEG mobilities at the given TD density. We therefore suggest that the achieved 2DEG mobilities of $>1750$ cm$^2$/V s (at 300 K) were dominated by alloy or interface scattering, rather than by reaching the mobility limit via charged dislocation scattering. The latter becomes typically much more dominant in the low sheet carrier density regime, i.e., at $n_{th} < 3 \times 10^{12}$ cm$^{-2}$, as calculated for the TD densities of $\sim 5 \times 10^{8}$ cm$^{-2}$ in our AlGaN/GaN layers.\cite{48} According to these numbers, we would expect significantly higher mobilities for sheet carrier densities adjusted to the low-to-mid $10^{12}$ cm$^{-2}$ region.

V. CONCLUSION

This work addressed several critical issues related to the PAMBE regrowth of AlGaN/GaN HEMT structures on (0001) GaN templates contaminated with typical residual impurities (O, C, and Si) at the RI. Utilizing an optimized in situ thermal cleaning procedure consisting of thermal desorption and regrowth cycles at temperatures in the thermal decomposition regime of GaN (i.e., 780 °C), O and C impurities were effectively desorbed from the RI. This cleaning step preserved an overall very smooth surface morphology while exposing only shallow surface pits associated with the typical TDs piercing through the GaN template interface. Due to the very low vapor pressure of Si impurities, their tenacity and $n$-type conductive behavior at the RI could only be compensated by intentionally doped GaN:C buffer layers for proper isolation and ultimate reduction of buffer leakage. Further investigations of AlGaN/GaN device layers grown on top under the recently established high-$T$ PAMBE growth conditions yielded several promises for low background impurity with large flexibility in AlGaN growth window. While both the high-$T$ GaN and AlGaN layers seemed to incorporate fairly low levels of unintentional oxygen (similar to the widely used Ga-rich/low-$T$ conditions), the AlGaN surface morphology (structure and rms roughness) was found independent of the (Al+Ga)/N flux ratio, at least from N-rich/high-$T$ to intermediate Ga-rich/high-$T$ growth conditions. 300 K 2DEG mobilities and sheet resistances of such N-rich/high-$T$-grown AlGaN/GaN heterostructures were $>1750$ cm$^2$/V s and 295 $\Omega$/sq, respectively (at not yet optimized sheet carrier densities of $\sim 1 \times 10^{13}$ cm$^{-2}$), with maximum dc drain currents of $\sim 1.1$ A/mm at a +1 V bias. We expect that with further adjustments of the sheet carrier density, much higher electron mobilities can be achieved toward the limit where charged dislocation scattering will become dominant within these fairly low TDD AlGaN/GaN heterostructures.

ACKNOWLEDGMENTS

The authors thank Charles Evans Analytical Group and Dr. T. Mates (UCSB) for SIMS measurements and Dr. David Storm, U.S. Naval Research Laboratories for helpful discussions. This work was supported by USAFOSR (Kitt Reinhardt, Program Manager) and DOE SSL Project No. DE-FC26-06NT42857. Further support was provided by ONR through the DRIFT MURI (Dr. Paul Maki, Program Manager).
