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Influence of Ga/N ratio on morphology, vacancies, and electrical transport in GaN grown by molecular beam epitaxy at high temperature

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The effect of Ga/N flux ratio on surface morphology, incorporation of point defects and electrical transport properties of GaN films grown by plasma-assisted molecular beam epitaxy in a recently developed high-temperature growth regime was investigated. The homoepitaxial (0001) GaN films grown at ~780–790 °C showed smoothest morphologies near the cross-over between N-rich and Ga-rich growth ($0.75 < Ga/N < 1.1$) contrasting previous observations for low-temperature growth. The higher-quality growth near $Ga/N \sim 1$ resulted from lower thermal decomposition rates and was corroborated by slightly lower Ga vacancy concentrations [V_{Ga}], lower unintentional oxygen incorporation, and improved electron mobilities. The consistently low [V_{Ga}], i.e., $\sim 10^{16}$ cm^{-3} for all films attribute further to the significant benefits of the high-temperature growth regime. © *2010 American Institute of Physics*. [doi:10.1063/1.3514236]

Progress in group-III nitride electronic devices has been largely aided by significant materials improvements using the versatile molecular beam epitaxy (MBE) growth technique.[1–3] Plasma-assisted (PA)-MBE based on rf activated atomic nitrogen (N) has traditionally enabled high quality GaN growth at fairly low temperatures (T ~ 650–740 °C) under kinetically limited group III-rich conditions.[4] In low-T MBE-grown GaN, impurity incorporation, point defect densities, and electrical properties,[5] were found extremely sensitive to growth stoichiometry (Ga/N flux ratio), i.e., both oxygen (O) and Ga vacancy (V_{Ga}) densities [mainly as V_{Ga}-donor impurity complexes, $V_{Ga}-O_N$ (Ref. 6)] were much higher in N-rich as in Ga-rich grown films.[7–9] These $V_{Ga}-O_N$ complexes are acceptor states in unintentionally doped (UID) n-type GaN and cause electrical compensation.[6,10–12]

Optimized films by Ga-rich/low-T PAMBE growth have, however, suffered from extreme growth control at the boundary for Ga droplet formation, and second, relatively large reverse-bias leakage currents through charged threading dislocations (TDs).[13] Recently, we surpassed these problems by growth at much higher temperatures [i.e, region of thermal decomposition (>760 °C)] and at surprisingly low Ga/N ratios ($Ga/N \leq 1$).[14–16] The major benefits were much wider growth window for smooth GaN without excess metallic Ga nor additional defects, yielding the highest ever reported bulk room temperature (RT) electron mobilities (>1150 cm^2/V s).[15,17] In addition, these conditions resulted in ultralow reverse bias leakage,[18] in significant contrast to low-T grown GaN films. Despite these advances systematic studies of the effect of the Ga/N ratio on surface kinetics, native point defects, and electrical transport within this promising growth regime are still missing.

Resolving these critical properties was the aim of this letter, using atomic force microscopy (AFM), secondary ion mass spectroscopy (SIMS), positron annihilation spectroscopy (PAS) and Hall effect measurements for a set of high-T homoepitaxial (0001) GaN films.

The GaN films were grown in a Gen-II MBE system using effusion cells for Ga, and Si and a Veeco Unibulb radio frequency plasma source for active nitrogen. All high-T GaN films were ~0.8–1 μm thick, lightly doped with Si (low-10^{16} cm^{-3} region), and grown at constant T ~ 780–790 °C but variable Ga/N flux ratio. Details on source purity, flux, and growth temperature calibration as well as underlying buffer layer structure are reported elsewhere.[15] The PAS experiments were performed at RT with a variable-energy positron beam and high purity Ge detectors.[19]

As substrates highly resistive (0001) GaN templates (Lumilog) with a TD density (TDD) of $\sim 5 \times 10^8$ cm^{-2} were used. For the PAS investigations, we compared films grown on these GaN templates also with films on lower TDD GaN substrates, i.e., (i) on epitaxial lateral overgrowth (ELO) (0001) GaN with an average TDD $\sim 4 \times 10^7$ cm^{-2} and (ii) on freestanding (0001) GaN with TDD $\sim 5 \times 10^6$ cm^{-2}.

Representative AFM morphologies for the high-T GaN films grown under different Ga/N ratios (range of 0.6–2.8) are shown in Fig. 1. The surfaces changed from terracelike structures (with multiple steps of *c* constant) under very Ga-rich conditions [Figs. 1(a) and 1(b)] to much smoother layer-by-layer-like morphologies under low Ga-rich and slightly N-rich growth [Figs. 1(c)–1(e)], and to increasingly pitted surfaces under more N-rich conditions [Fig. 1(f)]. Concurrently, the root-mean-square (rms) roughness over the selected 10×10 μm^2 area changed from 3.6 nm to 2.1, 0.8, 1.0, 1.2, and 3.8 nm consecutively from Figs. 1(a)–1(f).

While the layer-by-layer features in Figs. 1(c)–1(e) were similar to those found earlier for N-rich/high-T growth,[15] the terracelike structure under more Ga-rich conditions was opposed to the hexagonal spiral hillocks common under Ga-rich/low-T conditions.[5] The absence of spiral growth hillocks indicates an increased critical radius of curvature for two-dimensional (2D) island formation or spiral initiation, i.e., changes in step energy.[4,5] Most striking, the tendency for the smoothest surfaces near the cross-over from N-rich to Ga-

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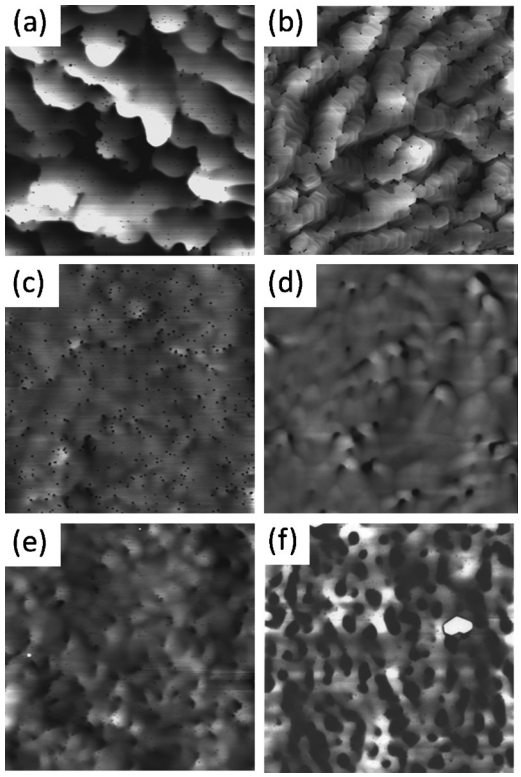

FIG. 1. 10×10 μm^2 AFM images of high-T PAMBE GaN films grown on standard (0001) GaN templates (TDD$\sim5\times10^8$ cm^{-2}) at fixed T=780 °C and N flux of 5 nm/min but variable Ga/N flux ratio of (a) 2.8, (b) 1.6, (c) 1.1, (d) 0.9, (e) 0.8, and (f) 0.6. The height scale is 10 nm.

rich growth (Ga/N$\sim$1), is in complete difference to the required Ga-rich/near-Ga-droplet conditions (Ga/N$\gg$1) for high-quality GaN at low-T (<750 °C).[4] We attribute this discrepancy to the modified surface growth kinetics under the influence of thermal decomposition at the high-T conditions,[13,20] as discussed below. On the other hand, the more pitted surface for the very N-rich grown film [Fig. 1(f)] was associated with increased excess active N on the surface leading to larger surface diffusion barrier.[4,21]

We note that light Si doping ([Si] $\sim10^{16}$ cm^{-3}) had no influence on the surface kinetics, as confirmed by qualitatively similar morphology of undoped GaN films grown under identical conditions. Thus, the N-rich/ high-T growth did not result in antisurfactant behavior by Si, as found for highly Si-doped N-rich grown GaN films at lower T.[22]

To identify the relation between Ga/N ratio, morphology and predominant point defects we used PAS to determine [V_{Ga}] from typical positron-electron pair annihilation characteristics.[19] The PAS measurements were conducted under different positron implantation energies in the 0–25 keV range while probing the positron trapping at Ga vacancies via Doppler broadening of the annihilation radiation line at 511 keV. The data were analyzed using the conventional *S* and *W* parameters, representing the fractions of annihilations with low and high momentum electrons.[19]

The *S* parameter measured as a function of positron implantation energy (mean implantation depth) is shown in Fig. 2 for selected high-T GaN films in reference to MBE grown p-type GaN, where positrons annihilate only in the GaN lattice.[10] For low-energy positrons (0–10 keV) implanted near the sample surface ($\sim$0.25 μm thick surface region) the *S* parameters were not constant ($0.48<S<0.44$). With increasing positron energy ($E>10$ keV) the positron diffusion to the surface decreased and the $S(E)$ curve saturated at constant levels of $S\sim0.44-0.45$, characteristic for the interior of the high-T GaN films.[10,19] The *S* parameters were all higher than the respective value for the reference p-GaN sample, indicating that vacancy-type defects are present in the high-T GaN films. Analysis within the region of interest (sample depth of $\sim$0.25–0.8 μm) shows that these vacancy defects have a slightly higher density in the very Ga-rich grown films (Ga/N$\gg$1) and reduced density with decreasing Ga/N ratio (Ga/N$\leq$1). Moreover, the high-T GaN film grown on the ELO GaN template (TDD of $\sim4\times10^7$ cm^{-2}) exhibited *S* parameter very close to that for the perfect GaN lattice.

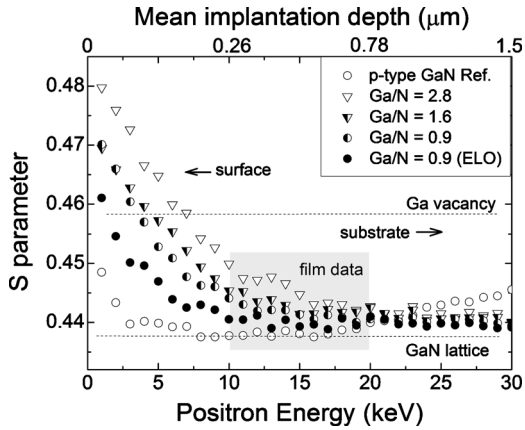

FIG. 2. *S* parameter measured as a function of positron implantation energy for high-T GaN films grown on both standard (0001) GaN templates (TDD$\sim5\times10^8$ cm^{-2}) and ELO-GaN template (TDD$\sim4\times10^7$ cm^{-2}) in reference to defect-free p-type GaN (as of Ref. 10).

Further, the *W* parameter was determined and plotted against the *S* parameter (*S*-*W* plot not shown), evidencing the typical linearity of valence and core electron momentum distributions. This demonstrated that the positrons annihilated from two different states and were identified as delocalized state in the perfect GaN lattice and the localized state at the in-grown V_{Ga} [11] allowing us to evaluate the absolute V_{Ga} density. By applying the positron trapping model,[19] [V_{Ga}] in the GaN films varied thus from $\sim2-6\times10^{16}$ cm^{-3}, with the lowest [V_{Ga}] near or slightly below Ga/N$\sim$1 [Fig. 3(a)]. Growth on lower-TDD GaN substrates at fixed Ga/N ratio of $\sim$0.9 (N-rich growth) yielded slightly lower [V_{Ga}] close to the detection limit of $\sim1\times10^{16}$ cm^{-3}. Considering the significant change in TDD, this decrease in [V_{Ga}] is negligible, indicating that V_{Ga} does not depend much on the presence of dislocations.[23] Overall, the values for [V_{Ga}] reported here are very low, similar to the typical [V_{Ga}] observed in highest-quality hydride vapor phase epitaxy GaN (Ref. 10) or Ga-rich/low-T-grown MBE-GaN (Ref. 7).

Analyzing the influence of Ga/N ratio on electrical transport, the RT Hall mobilities μ_e and respective bulk electron concentrations n_{bulk} for selected high-T GaN films are shown in Fig. 3(b). Consistent with the higher-quality morphology and lower [V_{Ga}], the highest μ_e (i.e., >870 and >1125 cm^2/V s) were achieved under slightly N–rich conditions for growth on standard and ELO-GaN templates respectively. Decreased μ_e at the more N-rich (Ga/N$\ll$1) and Ga-rich (Ga/N$\gg$1) sides was accompanied by slightly increased n_{bulk} from low-10^{16} to 1×10^{17} cm^{-3}. Since negatively charged V_{Ga} usually provide an acceptor in GaN, the

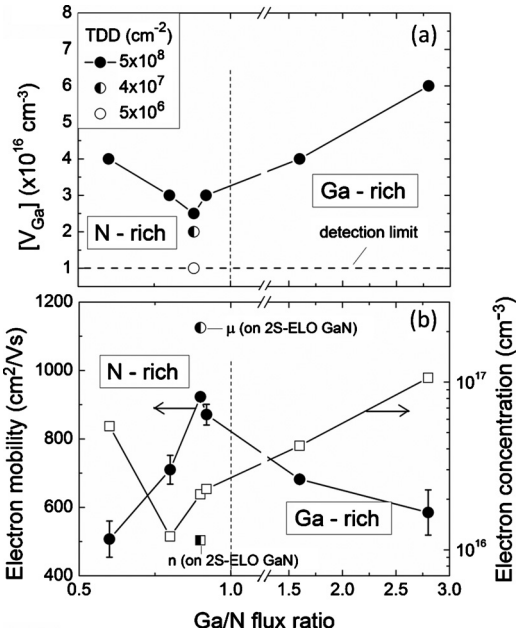

FIG. 3. (a) Calculated Ga vacancy concentration [V_{Ga}] as function of Ga/N ratio for the high-T GaN films grown at fixed T=780 °C and for different TDD-GaN substrates; (b) RT electron mobility and n-type carrier concentration for the same films as measured by Hall effect in *vdP* geometry.

increase in n_{bulk} must therefore arise from impurities such as (O) or (Si), which are predominant sources for n-type conductivity in GaN.[24] Based on SIMS measurements, the doped high-T GaN region showed only small variation in [Si], i.e., 2×10^{16}–6×10^{16} cm^{-3}, consistent with nominally supplied Si doping. On the other hand, UID [O] was in the low-10^{16} cm^{-3} region for GaN films grown at Ga/N ~ 1, and increased more toward ~10^{17} cm^{-3} when deviating from flux stoichiometry. Apart from the typical inaccuracy of SIMS these O-related doping effects are marginal but could be responsible for the slight increase in n_{bulk}, and correlate with the increase of [V_{Ga}], since O forms high-binding energy complexes with V_{Ga} (Refs. 6 and 24) (evident in Fig. 3(a) and in agreement with previous PAS studies[10–12]).

Interpreting the relationship between morphology, [V_{Ga}] and electrical properties, we considered the influence of Ga/N ratio on TD defects). Under variable Ga/N ratio, however, neither the TDD nor the relative proportion of TD-type (screw- versus mixed-, or edge-type component) was changed in the GaN films.[18] This supports the notion that [V_{Ga}] is completely independent of TDD,[23] and that the TDs do not contain vacancy defects in their core.[25]

Thus, the trends for variable [V_{Ga}] and [O] most likely relate to differences in growth kinetics and associated surface morphology, through the competition between the forward (growth) and reverse (decomposition) reactions at high-T. Previous work demonstrated increased thermal decomposition rates for Ga/N > 1 as opposed to Ga/N ≤ 1.[20] This explains the increased rms roughness under Ga-rich conditions, since with higher Ga desorption rates the Ga surface reactivity is reduced. Assuming that decomposition occurs by vacancy formation,[26] this suggests further a higher rate for vacancy generation under Ga-rich conditions.

Besides, decomposition also occurs near structural imperfections (dislocations, kinks, or step edges)[26] and is furthermore responsible for autodoping of donors in GaN.[27] This may also explain the higher propensity for step-edge formation, more distinct open volume surface pits (i.e., at the termination of TDs), and slightly higher [O] and n_{bulk}, which were observed more clearly in Ga-rich/high-T GaN films. In contrast, with more suppressed decomposition, N-rich/high-T grown GaN films resulted in smoother layer-by-layer like surfaces and decreased [V_{Ga}]. This interesting finding is in contrast to the common notion of much larger [V_{Ga}] in N-rich/low-T GaN,[23,27] and can be attributed to the much higher-quality growth under high-T growth conditions.

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