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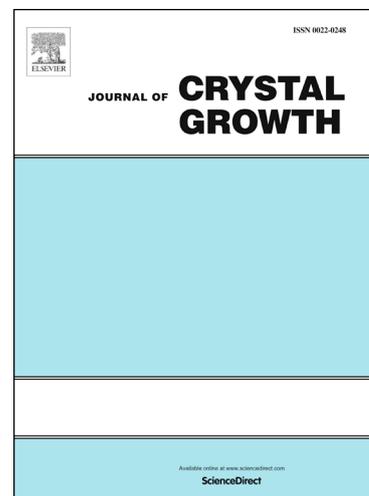
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Effect of substrate misorientation on the concentration of impurities and surface morphology of an epitaxial GaN layer on N-polar GaN substrate by MOVPE

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Abstract

This study examines the effect of (000-1) GaN substrate misorientation on the residual impurities and surface morphology of N-polar GaN grown by metalorganic vapor-phase epitaxy. Carbon, silicon, and oxygen concentrations decreased with increasing GaN substrate misorientation angle, with the lowest impurity concentration achieved for a misorientation angle of 2° toward the m-axis, with $6 \times 10^{15} \text{ cm}^{-3}$ carbon, $6 \times 10^{15} \text{ cm}^{-3}$ silicon, and $4 \times 10^{17} \text{ cm}^{-3}$ oxygen atoms. The oxygen concentration was measured at a depth of $0.5 \mu\text{m}$ below the wafer surface, and the oxygen concentration decreased with increasing thickness. The incorporation of carbon and oxygen revealed a strong dependence on the misorientation angle. The step distance height of the steps parallel to the [11-20] direction (or perpendicular to the [1-100] m-direction) was confirmed to be a double-height layer step. This phenomenon indicated that m-direction steps are stable for N-polar growth in GaN. In cases of large misorientation toward the m-axis in of the GaN substrate it was difficult to control the misorientation perpendicular to the nominal direction leading to a-axis direction by wafer bowing at wafer manufacturing. Therefore, step-bunching was generated for each symmetric m-axis due to an increase in the compound's off-angle, thus causing the surface roughness to become large.

Keywords: A1. Impurities; A1. Surfaces; A3. Metalorganic vapor-phase epitaxy; B1. Nitrides

1. Introduction

Nitride semiconductors have received increasing amounts of attention due to their application in high temperature power devices [1,2]. Mass production of AlGaN/GaN high-electron-mobility transistors, GaN-related power devices in the Ga-polar plane, has recently commenced [3,4]. GaN vertical power devices are being intensively studied using GaN substrates [5-8]. To achieve a breakdown voltage above 1 kV in unipolar GaN vertical devices, thicknesses exceeding 5 μm and carrier concentrations below $2 \times 10^{16} \text{ cm}^{-3}$ are required [9,10]. Unintentional carbon inclusions have resulted in the acceptor character of isolated substitutional C in the nitrogen site, generating discrepancies in the carrier concentration in the drift layer [11]. The high growth rate is attractive because growth time for drift layer can be reduced. However, the carbon concentration in GaN increases as the V/III ratio decreases for a high growth rate in the metalorganic vapor-phase epitaxy (MOVPE) [12]. N-polar GaN displays difficulty in the incorporation of carbon compared with Ga-polar from surface stability and growth kinetic perspectives [13]. Therefore, the carbon concentration in N-polar GaN is less than that of Ga-polar GaN, even under the same vapor-phase condition [13]. A much lower residual-carbon incorporation has been reported for MOVPE growth (000-1) GaN [13,14,15]. For vertical devices, the GaN substrate must also be conductive to avoid additional resistance between the substrate and the n-type layer. Thus, GaN growth in the (000-1) direction at the drift layer on GaN substrate in vertical power devices is generally expected to display better properties than in (0001) devices. For N-polar GaN, the surface morphology has been shown to significantly improve by increasing the off-cut angle on a strongly nitridated sapphire substrate [16-18]. Even under homoepitaxy, the N-polar GaN epitaxial layer displays many hillocks [19-21]. In cases of homoepitaxy, the effect of the misorientation angle on surface morphology and the concentrations of impurities has yet to be clarified. Therefore, we investigated the dependence of the impurity concentration and surface morphology of GaN on the misorientation angle of N-plane GaN substrates.

2. Experimental details

Crystal growth took place in a horizontal MOVPE reactor using a quartz-free flow channel. This channel is effective for reducing residual impurities of carbon and oxygen concentrations in an epitaxial m-plane GaN layer [22]. The susceptor configuration was 5×2 inches, and the growth temperature was $1,100^{\circ}\text{C}$ at 101.3 kPa of H_2 carrier gas. Trimethylgallium (TMGa) and ammonia (NH_3) were used as precursors. The TMGa flow rate was 700 $\mu\text{mol}/\text{min}$ with a V/III ratio of 1,019, and the growth time was one hour.

The N-polar GaN substrates were fabricated using hydride-vapor-phase epitaxy (HVPE) by Furukawa Electronics Co., Ltd. Table 1 lists the misorientation angles of GaN substrate in $[1-100]$ (m-axis) and $[11-20]$ (a-axis), as determined by X-ray diffraction analysis. The distribution of misorientation angles was less than $\pm 0.05^{\circ}$ in the 1×1 -cm wafer region. However, the orientation of the wafer became more difficult to control at higher misorientation angles, which is why the unintentional offset in the a-direction increased as the angle along the m-direction increased. The surface was epi-ready, which is similar to the treatment of the Ga-polar surface. Figure 1 shows an atomic force microscope (AFM) image of the typical surface morphology of an N-plane GaN substrate (here with a misorientation of 2.55° $_{[1-100]}$ and 0.32° $_{[11-20]}$). The root-mean-square (RMS) roughness value was 0.149 nm, and the N-polar surface after chemical polishing was very smooth without scratches, and there were no resolved atomic steps.

Table 1

sample	m-axis orientation [degree]	a-axis orientation [degree]
A	0.00	0.00
B	0.41	0.00
C	0.98	0.02
D	2.11	0.05
E	2.55	0.32
F	3.70	0.76
G	6.30	0.57

Table 1. The list of misorientation angles of the GaN substrate, as determined from XRD analysis

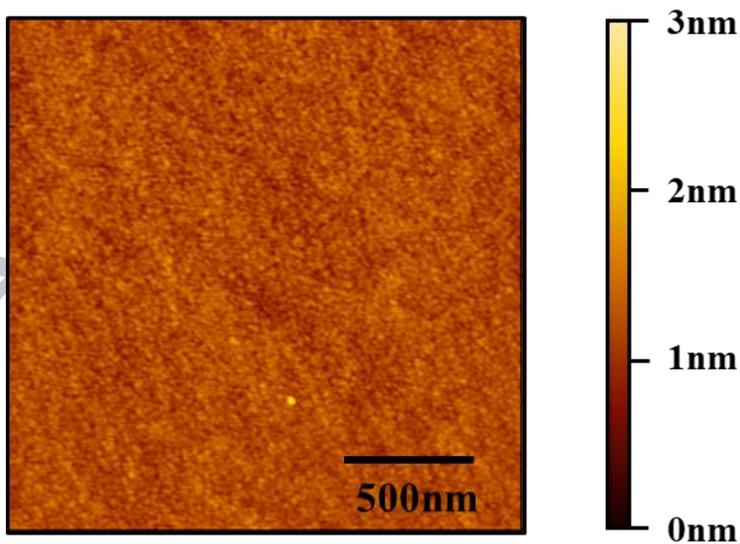


Figure 1. AFM image of the N-polar surface the GaN substrate with off-sets of 2.55° and 0.32° toward the m-axis [1-100] and a-axis [1-210], respectively.

The impurity concentration and growth rate of GaN on GaN were determined by secondary-ion-mass spectrometry (SIMS), and the surface morphology was measured with a Nomarski optical microscope and an AFM.

3. Results and discussion

3.1 Surface morphology of GaN

N-polar GaN layer growth by MOVPE is known to generate many hillocks. Zauner *et al.* reported an improvement in the surface morphology of N-polar GaN substrate [19,20], where surfaces dominated by hillocks were substantially reduced by increasing the misorientation angle from 0° to 4° off toward the m-axis, and a similar tendency was observed on the sapphire substrate [16-18].

Figure 2 shows the surface morphology of GaN grown on N-polar GaN substrate, as observed by a Nomarski optical microscope. Hexagonal hillocks were present in the sample grown on the m-axis 0° and 0.4° misorientation substrates, which is consistent with the finding in previous reports [19,20]. For misorientation angles between 1 and 2 degrees, the hillocks virtually disappear, and steps -bunching appears in parallel to the a-direction. For misorientations greater than 2.5° towards the m-direction, the misorientations along the a-direction became larger than 0.3° and large steps appeared along another m-direction due to step bunching (Fig. 2(e)). In case of the sample with the highest misorientation angle of 3.7° (Fig. 2(f)) the actual step-edge was rotated by oriented at about 60° to the m-direction misorientation (i.e., steps -bunching was again observed along the m-direction), as noted previously [18].

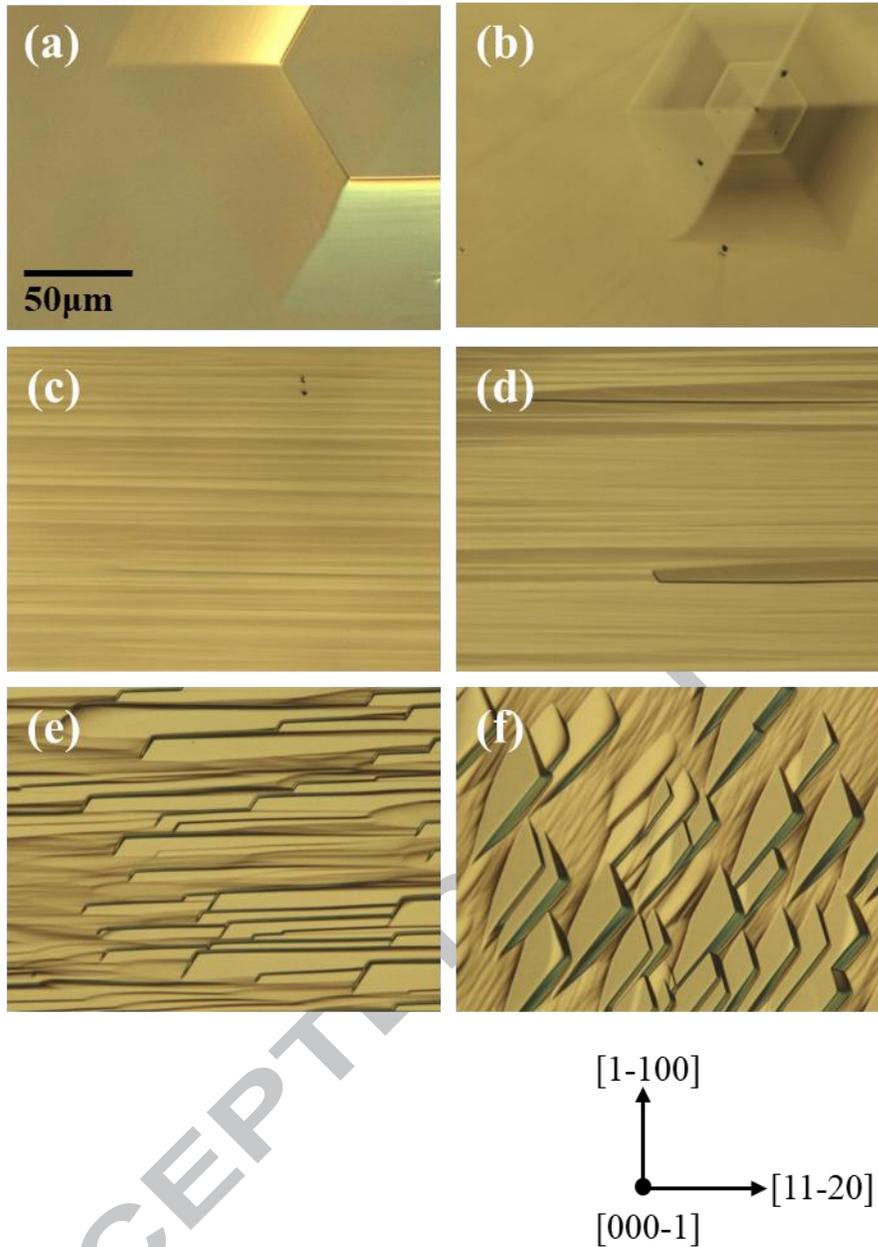


Figure 2. Differential-interference-contrast-microscopy images of N-polar GaN surfaces of misorientations along the m-axis of (a) 0°, (b) 0.4°, (c) 0.98°, (d) 2.11°, (e) 2.55°, and (f) 3.70°, respectively.

Figure 3 compares the misorientation dependency of hillock density and size with previous reports [18,20]. The trend revealed in this study was similar to that in both previous experiments, but hillock density in our sample was almost 100 times lower. According to their report, threading dislocations trapped Ga adatoms, which are thought to be the origin of the hillocks. Nowak *et al.* reported the relationship between hillock formation and dislocation [23]. We assume that this difference may originate either from the dislocation density of GaN substrate or from different MOVPE-growth conditions. Additionally, in our case, hillocks were eliminated for misorientation angles exceeding 1° .

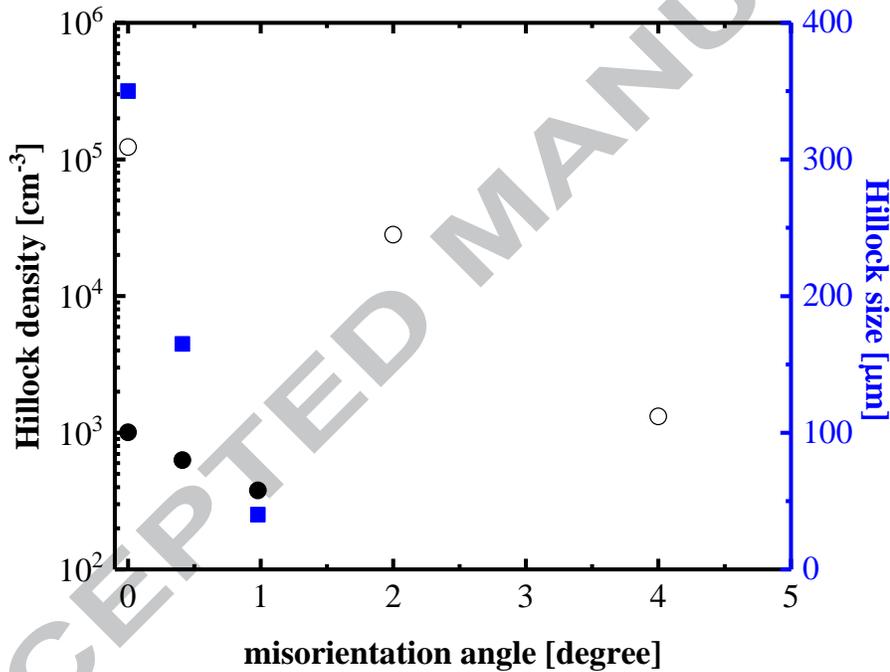


Figure 3. The misorientation angle dependence of hillock density and average hillock size

The hillock density is determined as the sum over a $900 \mu\text{m}$ by $700 \mu\text{m}$ area.

Blue square: hillock size in this experiment, Black circle: hillock density in this experiment, White circle: hillock density in previous reports [24,25]

Figure 4 shows a $2\ \mu\text{m} \times 2\ \mu\text{m}$ AFM image of the grown layer for various misorientation angles. The root-mean-square roughness (RMS) values of AFM were less than 0.5 nm at all orientation angles (i.e., they were limited by the AFM noise). For samples A-D, the steps clearly ran straight along the m-direction and kinks were observed. Only sample E did not follow the same trend as the step because high misorientation substrate along the m-axis was affected by the uncontrollable a-axis misorientation as indicated by the Nomarski microscope results (in Fig.2).

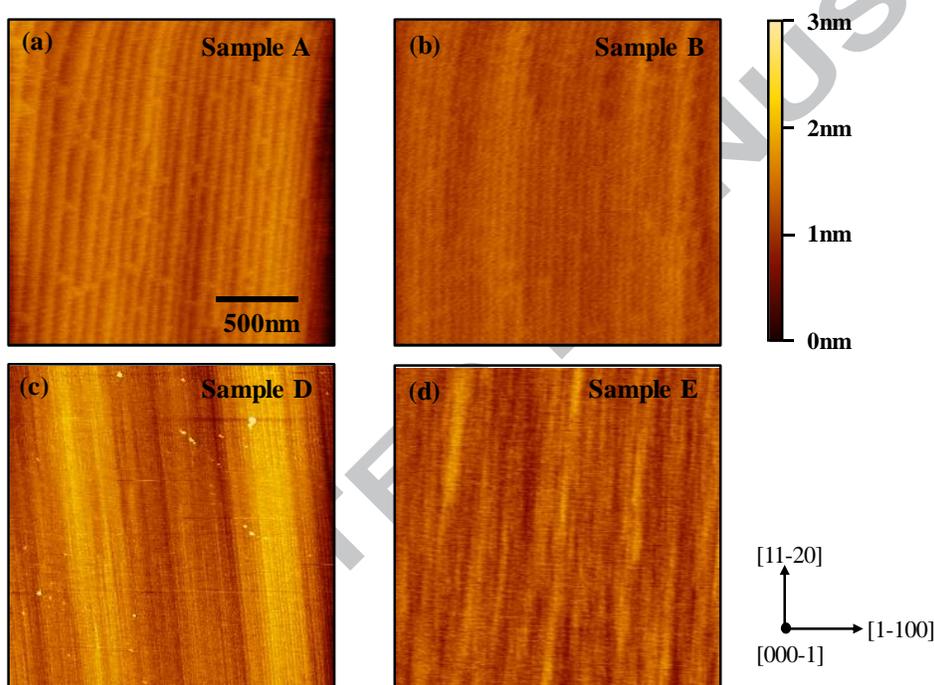


Figure 4. AFM image of a GaN epitaxial layer surface with different misorientation angles of (a) sample A, (b) sample B, (c) sample D, and (d) sample E. The measurement area and height are $2\ \mu\text{m} \times 2\ \mu\text{m}$ and 3 nm, respectively.

The step-to-step distance (assuming double-height steps [19]) was related to the misorientation of the observed terrace. However, this misorientation angle deviated from the actual misorientation angle as shown in Fig.5. The black line was calculated by assuming m-steps as suggested by the literature [19]

(i.e., double-atomic-layer steps). Indeed, the observed step length was close to the expected length, except for the 0° to 2.5° misorientations. In the latter case, the steps may be too close to be resolved by our AFM, and in the 0° case, the steps originated from the local misorientation at the measurement point. Hence, the results confirm the formation of double-atomic-height layer steps, which are also referred to as dominating m-direction steps parallel to the a-direction for crystal misorientations towards the m-direction.

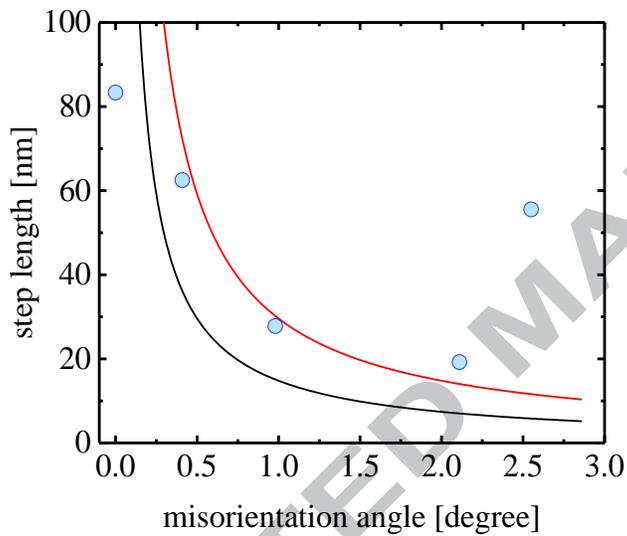


Figure 5. Length of the atomic step in GaN with misorientation dependence. The dots depict the experimental results obtained by AFM measurement. The black line is the theoretical value of step length in a single atomic layer, and the red line is the theoretical value of a double-layer height.

Apparently, the formation of m-type steps, and the resulting step-bunching prevents the formation of hillocks for misorientations larger than 1° (for which the corresponding terrace spacing was smaller than 30 nm). While the formation of hillocks can be suppressed by a misorientation in the a-direction, the resulting zig zag step-bunching (due to the m-steps being more stable) is not ideal. The

step-bunching of the resulting surface is affected by the unintentional misorientation along the a-direction as shown in Fig. 2e. Hence, when an inclined m-direction plane is used, the unintentional a-axis off-angle must be as small as possible. In this experiment, an a-axis off-angle of 0.32° was found not only to induce macro-sized step-bunching perpendicular to the misorientation direction, but also to the impact led to the formation of additional surface steps.

3.2 Growth rate and impurities concentration

In the previous section, the surface morphology's dependence on the misorientation angle was discussed. In this section, we examine the growth rate and concentrations of unintentional impurities. Detailed results regarding the incorporation of impurities under several growth conditions on the N-polar GaN on sapphire have previously been reported [24-26]. The growth rate also depends on the misorientation, as shown in Fig. 6. Sample-A had a high hillock concentration, and hillock affected the SIMS measurement. However, sample B-G had a delineated hillock-free area, which avoided the effect of hillock. For misorientations below 2° , the growth rate increased with increasing misorientation because of step length and surface morphology. The growth rate in the sample with a misorientation over 2° displayed a different trend compared with samples having misorientations below 2° because the other symmetric m-direction step-bunching appeared due to effect of the unintentional misorientation.

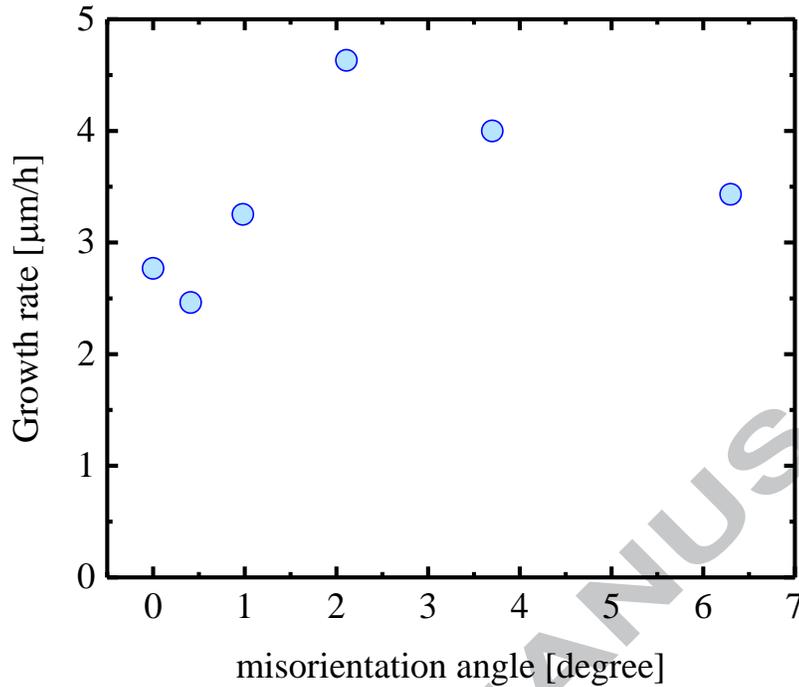


Figure 6. Growth rate of GaN with various misorientation angles. The growth rate of 4.7 $\mu\text{m/h}$ was achieved by sample D.

Figure 7 shows a typical oxygen concentration depth profile for our N-polar GaN, here for the 3.7° misoriented sample. The oxygen concentration decreases as the thickness increased for all misorientation angles. A similar trend was observed with m-plane GaN in just off-angle and +c-axis incline [15]. The origin of this trend is not clear, but it may be related to the coating of the sapphire dummy by GaN. However, the oxygen concentration in N-polar GaN was much higher than that of m-plane GaN or Ga-polar (0001) GaN.

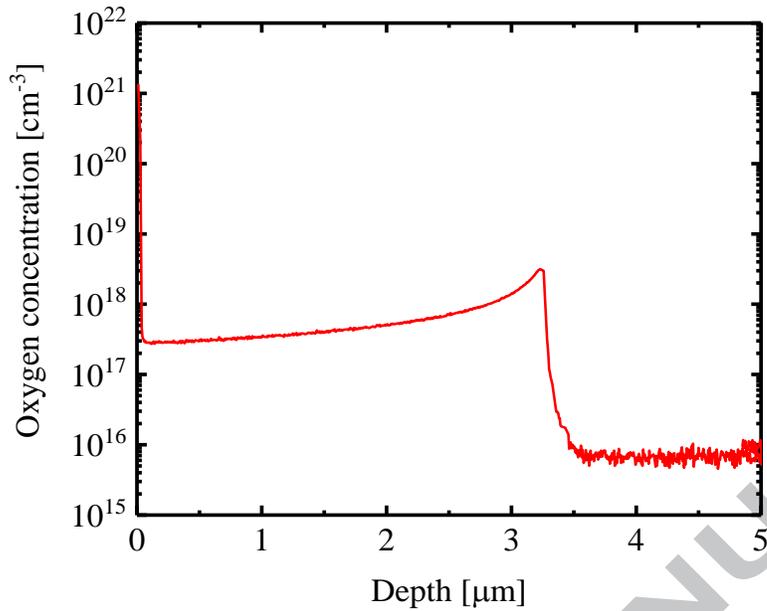


Figure 7. SIMS measurement of oxygen by sample F. The detection limit was $7 \times 10^{15} \text{cm}^{-3}$.

Figure 8 compares the carbon, silicon, and oxygen concentrations after 1 hour of growth of GaN as a function of the misorientation angle. The carbon and oxygen concentrations decreased as the misorientation angle increased and tended to saturate above 2° ; hence, the hillocks are favorable incorporation sites for carbon and oxygen, because they are composed of various stable semi-polar planes. [15, 27]. The silicon concentration is inversely proportional to growth rate, which is thought to be due to decomposition of silicon from the SiC coat flow channel due to the high growth temperature. The carbon, silicon, and oxygen concentrations in GaN grown on the Ga-polar (0001) GaN substrate were 1.2×10^{16} , 3×10^{15} , and $7 \times 10^{15} \text{cm}^{-3}$, respectively. Comparing these results to Fig. 8, the carbon concentration in N-polar (000-1) GaN at misorientation angles over 2° was less than $6 \times 10^{15} \text{cm}^{-3}$, and the concentration was less than half that for the Ga-polar GaN.

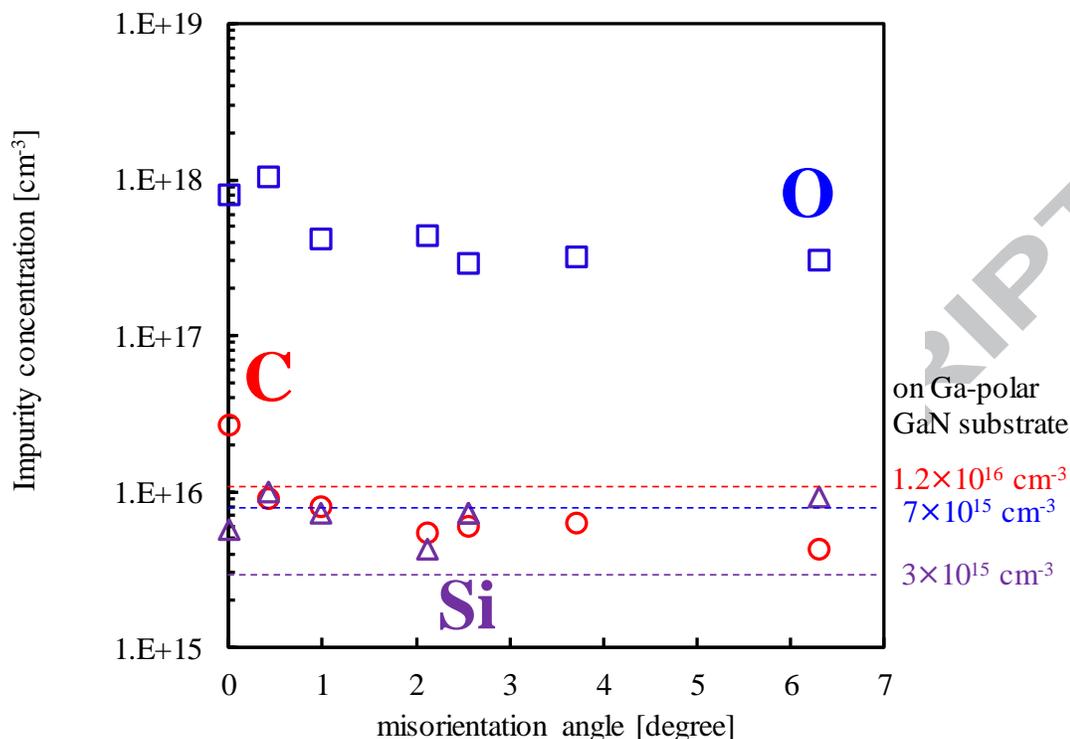


Figure 8. The misorientation angle dependences of the impurity concentrations of carbon, silicon, and oxygen. These measurement detection-limits for carbon, silicon and oxygen are $3 \times 10^{15} \text{ cm}^{-3}$, $1 \times 10^{15} \text{ cm}^{-3}$, and $7 \times 10^{15} \text{ cm}^{-3}$, respectively. The oxygen concentration is determined 0.5 μm from the surface. The dotted line represents the results of the Ga-polar plane GaN with substrate misorientations of 0.03 and 0.4 toward the m-axis [1-100] and a-axis [1-210], respectively. The red, violet, and blue dots are the carbon, silicon, and oxygen concentrations.

N-polar GaN has a much higher oxygen concentration, but its carbon concentration was quite low. To reduce the oxygen concentration, each material should not only use a quartz-free flow channel, but also a dummy substrate and raw materials. In any case, the growth rate of 4.7 $\mu\text{m}/\text{h}$ with a carbon concentration of only $5 \times 10^{15} \text{ cm}^{-3}$ (sample D) is very encouraging.

4. Conclusion

We studied the impact of misorientation on the homo-epitaxial growth of (000-1) GaN. Using a misorientation above 1° along the m-direction and avoiding any misorientation along the a-direction, a smooth, hillock-free surface was obtained with a high growth rate and low carbon concentration. To avoid zigzag step-bunches, the a-direction misorientation should be controlled to within 0.3° . This indicates that N-polar GaN on a GaN substrate is a promising candidate for use in vertical power devices. However, the oxygen concentration of N-polar GaN grown on the GaN substrate is still higher than that of Ga-polar GaN. This oxygen concentration must be reduced to less than $1 \times 10^{16} \text{ cm}^{-3}$ if GaN is to be used as a drift layer in vertical power devices.

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Highlights

1. Low impurities concentration in GaN grown by MOVPE using quartz free reactor
2. N-polar GaN atomic steps with double height.
3. Different off-cut angle N-polar GaN for homo-epitaxial growth.