Correlation between nanopipes formed from screw dislocations during homoepitaxial growth by metal-organic vapor-phase epitaxy and reverse leakage current in vertical p–n diodes on a free-standing GaN substrates

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We fabricated p–n diodes under different growth pressures on free-standing GaN substrates of the same quality and observed a noteworthy difference in the reverse leakage current. A large reverse leakage current was generated by nanopipes, which were formed from screw dislocations in the homoepitaxial layer. There were two types of screw dislocation observed in this study. The first type already existed in the substrate and the other was newly generated in the epilayer by the coalescence of edge and mixed dislocations. An increase in the growth pressure suppressed the transformation of screw dislocations into nanopipes, which led to a reduction in the reverse leakage current. To reduce the leakage current further, it is necessary to apply growth conditions that do not transform screw dislocation into nanopipes and to use a free-standing substrate without threading dislocations, that become nanopipes. © 2019 The Japan Society of Applied Physics

1. Introduction

GaN is an attractive material for next-generation power semiconductor devices owing to its high critical breakdown electric field, high electron mobility, and high electron saturation velocity. In recent years, vertical GaN devices homoepitaxially grown on high-quality, free-standing GaN substrates with low threading dislocation density (TDD) have been reported. They have an extremely low leakage current compared with GaN devices grown on foreign substrates and have an extremely low leakage current in vertical p–n diodes owing to its high critical breakdown electric field. GaN is an attractive material for next-generation power devices.1–10

In homoepitaxial growth, a dislocation in a free-standing substrate can propagate to the epitaxial layer and penetrate to the surface unless it is annihilated by forming a dislocation half loop. Therefore, there is a concern that the device yield may be reduced in large devices intended for high-current applications. Screw, mixed, and edge dislocations are types of threading dislocation (TD) found in GaN.11–15 There are two types of screw dislocation: hollow-core and full-core dislocations. A hollow-core screw dislocation is called a nanopipe, whose core diameter is typically about 10–50 nm.16–20 In this paper, a full-core screw dislocation is called simply a screw dislocation. The effect of these TDs on devices has been reported by several groups, and it is well known that in Schottky barrier diodes, reverse leakage occurs at screw dislocations and nanopipes.21–25 Leakage due to screw dislocations was confirmed in a p–n diode as well as in a Schottky barrier diode.26 However, inconsistent behavior has been reported for nanopipes. There are contradictory reports on whether leakage occurs at nanopipes in p–n diodes.26–28 Therefore, the relationship between TDs and leakage in p–n diodes is still controversial. For GaN-based power semiconductors with a high TDD, improving the yield for large devices is an urgent issue; thus, it is indispensable to identify the types of TD that cause a leakage current.

Since a dislocation can penetrate through an epilayer from a substrate, the device yield may be limited by the screw dislocation or nanopipe density of the free-standing substrate. However, we have found that the device yield varies significantly with the growth pressure in metal-organic vapor-phase epitaxy (MOVPE), even if free-standing GaN substrates of the same quality are used. This indicates that it may be possible to inactivate the TDs that result in a leakage current. In this study, we systematically investigated the mechanisms behind the MOVPE growth pressure dependence of the device yield and we report the effect of nanopipes on the electrical properties of vertical p–n diodes.

2. Experimental methods

2.1. Fabrication of vertical p–n diodes

A schematic cross section of a p–n diode is shown in Fig. 1. Specifically, a 200 nm n-type GaN layer (Si, 3 × 1018 cm−3), a 13 μm n-type drift layer (Si, 2 × 1016 cm−3), a 600 nm p-type GaN layer (Mg, 4 × 1019 cm−3), and a 30 nm p+ GaN contact layer (Mg, >1 × 1020 cm−3) were epitaxially grown by MOVPE on an n-type free-standing GaN substrate. The p–n diode structure was grown under three different pressures: 500 hPa (low pressure), 750 hPa (intermediate pressure), and 1000 hPa (atmospheric pressure). They are abbreviated here to LP-PND, MP-PND, and AP-PND, respectively.

The free-standing substrates used for each growth pressure are available commercially. The same substrate fabrication method and substrates from the same vendor were used to ensure the same quality of the free-standing GaN substrates.
The reverse $I-V$ characteristics were measured for 10 randomly selected LP-PNDs, MP-PNDs, and AP-PNDs with an electrode diameter of 100 $\mu$m. Then the device yield was evaluated.

The leakage spots were observed with an emission microscope (Hamamatsu Photonics PHEMOS-1000) using p–n diodes with an electrode diameter of 100 or 500 $\mu$m to investigate the origin of the leakage current. The emission microscope captures emission from visible light to the near infrared (400–1100 nm) using a Si-CCD camera. It detects a leakage spot as a light emission spot.\(^{30,31}\)

After observing the leakage spots, the etch pit method was applied to confirm the correlation between leakage spots and TDs. We used molten KOH as the etchant. The insulator and the electrode were dissolved before forming the etch pits. Since it was difficult to form the etch pits on the p-layer, the p-layer was removed by ICP-RIE and etch pits were formed on the exposed drift layer (the light blue region in Fig. 1). The KOH etching performed at 450 °C for 1 to 3 min By comparing the positions of leakage spots and etch pits, we could determine whether the leakage spots were due to TDs. It is known that the shape of an etch pit reflects the TD type.\(^{17,32–38}\) Thus, the etch pit types and their densities were compared among LP-PNDs, MP-PNDs, and AP-PNDs to confirm whether the proportion of each TD type changed during the growth.

To identify the TD type causing a reverse leakage current, TD propagation under the pits was first observed with a two-photon excitation microscope (Nikon, A1MP series). The microscopic observation of the two-photon excitation under the pits was carried out as follows. First, the diode with p-GaN removed was etched in molten KOH at 350 °C for 30 s to form etch pits as small as 1 $\mu$m in diameter to prevent scattering of the incident laser. Next, a Ti–sapphire laser with a wavelength of 700 nm was focused and scanned in the XY direction on the sample, and a two-dimensional (2D) intensity mapping image of two-photon excitation photoluminescence (2PPL) was acquired using a 2PPL system. As the 2PPL intensity around the TDs decreases, the TDs are observed as dark spots. Three-dimensional TD propagation was observed by stacking the contrast-inversed 2D mapping images obtained at each Z position while changing the focus position in the Z direction. For details of the 2PPL system, see Ref. 39.

Since the propagation direction of the TD under a pit was known from the 2PPL observation, the orientation of the cross section STEM specimen cut by a focused ion beam (FIB) was selected to include the entire dislocation under the pit. This allowed the entire TD under the pit to be analyzed by cross-sectional STEM. The TD type was determined by the invisibility criterion for the TD type.\(^{17,32–38}\) Thus, the TD types and their densities were comparing the positions of leakage spots and etch pits, we could determine whether the leakage spots were due to TDs. It is known that the shape of an etch pit reflects the TD type.\(^{17,32–38}\) Thus, the etch pit types and their densities were compared among LP-PNDs, MP-PNDs, and AP-PNDs to confirm whether the proportion of each TD type changed during the growth.

3. Results and discussion

3.1. Correlation between leakage and dislocations

The measured reverse $I-V$ characteristics and device yield of LP-PNDs, MP-PNDs, and AP-PNDs with electrode diameters of 100 $\mu$m are shown in Fig. 2. All the devices underwent nondestructive breakdown at ~900 V. To evaluate the device yield, we defined a device that exhibits a leakage current of less than 10$^{-6}$ A cm$^{-2}$ at ~850 V as a good product. The threshold point is shown by a star ($\star$) in Fig. 2. The device yield significantly increased from 30% for the
LP-PNDs to 90% for the AP-PNDs as the growth pressure increased. All the good and bad products in LP-PNDs were observed by the emission microscope, and representative emission images are shown in Fig. 3. At least one dotlike leakage spot was observed in all bad products while no leakage spots were observed in good products. This means that the origin of these leakage spots causes the large leakage current. The leakage spot densities of the LP-PNDs, MP-PNDs, and AP-PNDs were calculated by averaging the result for 10 devices with an electrode diameter of 500 μm.

Figure 4(a) shows a typical emission image of each p−n diode. The average leakage spot density for each growth pressure was $4.8 \times 10^3$, $1.5 \times 10^3$, and $9.2 \times 10^2$ cm$^{-2}$ for the LP-PNDs, MP-PNDs, and AP-PNDs, respectively. The leakage spot density decreased with increasing growth pressure. This reduction in the leakage spot density is consistent with the improvement in the device yield shown in Fig. 2.

After removing the insulator, electrode, and p-type layer, etch pits were formed on the exposed drift layer with molten KOH at 450 °C for 1–3 min. Figure 4(b) shows an optical microscope (OM) image of an LP-PND, MP-PND, and AP-PND after etch pit formation. From the OM images, three sizes of etch pit can be observed (large, medium, and small).
Figure 4(c) shows scanning electron microscopy (SEM) images of each etch pit type. The densities of the large, medium, and small pits in the LP-PNDs, MP-PNDs, and AP-PNDs were determined by averaging the number of each etch pit type in 10 devices with a mesa diameter of 540 μm. The density of each etch pit type in LP-PNDs, MP-PNDs, and AP-PNDs was summarized in Table II. The density of small pits was almost constant at around $3 \times 10^6 \text{cm}^{-2}$ for each growth pressure. The density of medium pits did not depend on the growth pressure. However, focusing on the density of large pits, it decreased by one order of magnitude from $1.4 \times 10^4 \text{cm}^{-2}$ for the LP-PNDs to $1.2 \times 10^3 \text{cm}^{-2}$ for the AP-PNDs as the growth pressure increased. When the leakage spot positions were superimposed on the OM images, as shown in Fig. 4(b), it was revealed that some of the large pits coincided with the leakage spots. The proportion of leakage spots corresponding to the large pits among all the leakage spots in the LP-PNDs, MP-PNDs, and AP-PNDs was calculated. We defined this proportion as the “matching rate of large pits,” which is also summarized in Table II. 94%, 70%, and 60% of leakage spots were matched with large pits in LP-PNDs, MP-PNDs, and AP-PNDs, respectively. This means that dislocations forming large pits are very likely to be the origin of the leakage spots. Therefore, it is considered that the reduction of the large-pit density due to the increase in the growth pressure lowers the leakage spot density, and consequently the device yield improves.

The medium pits did not coincide with the leakage spots. The dislocations forming medium pits were not responsible for the reverse leakage. There were leakage spots that did not match the large and medium pits. These leakage spots may have been caused by dislocations that form small etch pits, or other defects induced during the device fabrication process or the epitaxial growth.

Although some large pits coincided with the leakage spot, there were large pits that did not coincide with leakage spots. Generally, the size of the etch pit reflects the TD type. Therefore, there should be the same TD type under the large pits. This indicates that TDs of the same type may have different electrical properties.

### 3.2. Dislocation propagation under large pits

The TDs that formed large pits were one of the causes of the reverse leakage current and affected the device yield. Before performing cross-sectional STEM analysis on the TD type of large pits, dislocation propagation was observed using the 2PPL system. For the 2PPL observation, all the large pits should have a diameter of around 1 μm to avoid scattering of the incident laser. Figure 5(a) shows the shape of a large pit formed at $350 \degree C$ for 30 s in molten KOH. The diameter of the pit was around 1 μm. Although its shape is different from that of the large pit in Fig. 4(c), when it was additionally etched at $450 \degree C$ for 1 min, its shape became the same as shown in Fig. 5(b). Therefore, the etch pits with a shape similar to that in Fig. 5(a) formed at $350 \degree C$ for 30 s are the large pits.

The dislocation propagation was observed under 36 large pits that corresponded to leakage spots (“large pits with leakage”) and 39 large pits that did not correspond to leakage spots (“large pits W/O leakage”) in an LP-PND. Seven large pits with leakage in an AP-PND were also observed to analyze the growth pressure dependence of the dislocation propagation behavior. Owing to the low large-pit density, we could not increase the number of large pits observed in the AP-PND.

Observing the TD propagation, two kinds of propagation behaviors were observed under both the large pits with leakage and large pits W/O leakage. One is a coalesced-type, in which adjacent TDs merged in the epilayer and became a dislocation which formed large pit, and the other

| Growth pressure (hPa) | Large-pit density (cm$^{-2}$) | Medium pit | Small pit | Leakage spot density (cm$^{-2}$) | Matching rate of large pit |
|----------------------|-------------------------------|------------|-----------|-------------------------------|--------------------------|
| 500                  | $1.4 \times 10^4$             | $9.9 \times 10^2$ | $\sim 3.0 \times 10^6$ | $4.8 \times 10^1$ | 94%                      |
| 750                  | $4.0 \times 10^3$             | $1.3 \times 10^4$ | $\sim 3.0 \times 10^6$ | $1.5 \times 10^3$ | 70%                      |
| 1000                 | $1.2 \times 10^4$             | $6.0 \times 10^1$ | $\sim 5.0 \times 10^6$ | $9.2 \times 10^2$ | 60%                      |

![Fig. 5.](image) (Color online) OM and SEM images of large pits (a) etched at $350 \degree C$ for 30 s and (b) after additional KOH etching at $450 \degree C$ for 60 s. The shape of the etch pit became the same as that of the large pit shown in Fig. 4(c).

![Fig. 6.](image) (Color online) Image of dislocation propagation under large pit observed using 2PPL system. (a) and (b) Plan-view images of coalesced-type and single type, respectively. (c) and (d) Cross-section images of coalesced-type and single type, respectively, observed from the direction of the arrow shown in the corresponding plan-view image.
was a single type that penetrated the epilayer without coalescing and it formed also a large pit. Figure 6 shows the 2PPL observation results for the coalesced-type and single type under a large pit with leakage in an LP-PND. Figures 6(a) and 6(b) show plan views and Figs. 6(c) and 6(d) show cross-sectional views of each etch pit. The relationship between these two types of TD propagation and leakage is summarized in Table III. 69% and 100% of the large pits with leakage in the LP-PND and AP-PND were the coalesced-type, respectively. The coalesced-type tended to leak easily. If the growth pressure affects the dislocation propagation and suppresses the coalescence of TDs, the density of large pits with leakage will decrease and the yield will be improved. Therefore, we investigated the growth pressure dependence of the coalesced TD density. The density of coalesced TDs was determined by observing the propagation of more than 1400 TDs in LP-PNDs and AP-PNDs using the 2PPL system. It was $7.0 \times 10^4 \text{ cm}^{-2}$ for the LP-PNDs and $7.5 \times 10^4 \text{ cm}^{-2}$ for the AP-PNDs, which are almost the same. Therefore, the probability of TD coalescence does not depend on the growth pressure. The origin of the improved device yield is not the suppression of the coalescence of TDs but by other causes.

### 3.3. Dislocation types under large pits

Cross-sectional STEM analysis was performed to determine the dislocation type under the large pits. The following five kinds of large pit were observed as shown in Table III: the coalesced-type and single type large pit with leakage in LP-PNDs, the coalesced-type and single type large pit W/O leakage in LP-PNDs, and the coalesced-type large pit with leakage in AP-PNDs. The STEM images were observed under $g = [0002]$ and $g = [1120]$ conditions and are shown in Figs. 7–11. Since the orientation in FIB processing was determined by 2PPL observation, a cross-sectional STEM image of an entire dislocation penetrating the epilayer could be observed. Figures 7 and 8 correspond to Figs. 6(c) and 6(d), respectively.

A schematic cross section of the obtained STEM images is shown in Fig. 12. The coalesced-type in Figs. 7, 9, and 11 consisted of regions I to III and the single type in Figs. 8 and 10 consisted of regions I and II.
and 10 consisted of regions I and II. Region I is immediately under a large pit. All the STEM images showed two parallel dislocation lines in region I. To investigate this dislocation, region I of the AP-PND in Figs. 11 was reprocessed to obtain a plan-view STEM specimen, and the observed plan-view image is shown in Fig. 11(c). There was a hollow-core dislocation with diameter of 61 nm. The hollow-core dislocation in GaN is known as a “nanopipe” and only has a screw component \( (b = 1c, 2c, 3c \ldots) \).\(^{16,19,20,40} \) The plan-view of region I in an LP-PND was also observed and a nanopipe was also seen. This indicates that all the large pits were formed on nanopipes. We found that there were nanopipes causing a leakage current and nanopipes not causing a leakage current.

The nanopipes were formed from TDs propagating obliquely or meandering in the epilayer at point \( t \), and the TDs are shown with segment \( s \) in Figs. 7–11. The region before the transformation to the nanopipe was defined as region II. From the STEM and 2PPL observation results, the transformation of the TD from region II to region I did not involve the coalescence or splitting of the TD. From the conservation law of the Burgers vector, the dislocation in region II could be a screw dislocation. If a TD is a pure screw dislocation, its contrast should vanish under the \( g = [11\bar{2}0] \) condition. However, in the cross-sectional STEM image taken under the \( g = [11\bar{2}0] \) condition, residual contrast was observed in region II. This residual contrast may have been caused by the existence of basal edge dislocations. The screw dislocations propagating obliquely or meandering should include basal edge dislocations. For some orientation of the basal edge dislocation \( u, \mathbf{g} \cdot (\mathbf{b} \times \mathbf{u}) = 0 \) is not satisfied, resulting in residual contrast. In the enlarged views in Figs. 8(b) and 10(b), the dislocations propagating in a spiral show periodic contrast, which may reflect the change in orientation of the basal edge dislocations.

In the STEM images of the coalesced-type large pits in Figs. 7, 9, and 11, it was observed that two TDs (segments \( e \) and \( m \))
and m) coalesced and became one TD (segment s). The region before the coalescence is defined as region III. In the bright-field cross-sectional STEM images under both the $g = [1\bar{1}20]$ and $g = [0002]$ conditions, the dislocation indicated by segment m was observed as a dark line and was identified as a mixed dislocation. The dislocations in segment e appeared as dark lines under the condition $g = [1\bar{1}20]$ and showed residual contrast under the condition $g = [0002]$. The edge dislocation shows residual contrast because $g \cdot (b \times u) = 0$ is not satisfied under the condition $g = [0002]$ for some directions $u$. Therefore, the dislocation of segment e was identified as an edge dislocation.

3.4. Edge component in coalesced-type dislocation

In the coalesced-type dislocations in Figs. 7, 9, and 11, edge and mixed dislocations coalesced, and a dislocation indicated by segment s was formed. It was determined whether this dislocation was a screw dislocation or another dislocation by drawing the Burgers circuit to the edge and mixed dislocations before coalescence. If the edge components of both dislocations cancel each other upon coalescence, it is a screw dislocation, whereas it is a mixed (or edge) dislocation if the edge component remains.

The cores of TDs before coalescence under the coalesced-type large pits were observed by plan-view HAADF-STEM. The large pits observed in an LP-PND were large pit with leakage and large pit W/O leakage. Since these TDs are newly selected from the same LP-PND, they are different from the TDs observed by cross-sectional STEM in Figs. 7 and 9. The 2PPL images of the observed TDs are shown in Fig. 13. Similar to Figs. 6(a) and 6(c), it was observed that the two TDs coalesced and became one TD. The HAADF-STEM observation before coalescence was performed as follows: first, we picked-up the block of GaN including the large pit from the diodes by FIB in the determined orientation by 2PPL observation. Next, the picked-up samples were divided above and below the point where coalescence occurred. Finally, the part before coalescence was thinned for HAADF-STEM observation, and the part after coalescence was observed by plan-view STEM. A schematic image of this processing is shown in Fig. 14. The observed plan-view HAADF-STEM and STEM images before and after coalescence are shown in Figs. 15 and 16, respectively. As shown in Figs. 15(a) and 16(a), nanopipes were observed after coalescence similar to in the cross-sectional STEM observation. The nanopipes were too large for a Burgers circuit to be drawn. When we drew a Burgers circuit on the HAADF-STEM images of the dislocation pairs before coalescence, it was found that the 1a edge components were in opposite directions. This means the edge components will cancel each other as a result of the coalescence. Since the nanopipes were observed after coalescence, it was confirmed that they do not have edge components. Combining this with the result of cross-sectional STEM observation, it was clarified that dislocation s was a screw dislocation.

From these results, there were no difference between the large pits with leakage and large pits W/O leakage. Both were formed on the nanopipes formed from screw dislocations. However, the difference in the Burgers vector of both the screw dislocations before transformation is unknown from our study. Also, impurities on the nanopipe walls can affect the electrical properties. To clarify the origin of the leakage at nanopipes, further Burgers vector and impurity analyses are required.
All the nanopipes were formed from screw dislocations, and the probability of coalescence was not affected by the growth pressure. Therefore, it can be considered that the high growth pressure prevents the transformation from screw dislocations to nanopipes. This caused nanopipe (large pit) density to decrease with increasing growth pressure. As a consequence, the leakage spot density also decreased, this improving the device yield.

4. Conclusions
We investigated the mechanisms behind the improved device yield by increasing the growth pressure of a vertical p–n diode on a free-standing GaN substrate with a TDD of $3 \times 10^6$ cm$^{-2}$. We showed that a large leakage current is generated by nanopipes formed from screw dislocations and that a screw dislocation can be newly generated by the coalescence of an edge dislocation and a mixed dislocation in the epilayer. We found that the probability of TD coalescence does not depend on the growth pressure. Therefore, the device yield can be improved by suppressing the transformation of screw dislocations to nanopipes by increasing the growth pressure. To reduce the leakage current further and to improve the yield of large devices, it is necessary to apply optimized growth conditions in which screw dislocations are not transformed to nanopipes and to use a free-standing substrate without dislocations that are transformed into nanopipes.
There are nanopipes that cause leakage and nanopipes that do not cause leakage, which indicates that the TD type alone is not responsible for the leakage current and that there is another factor. We are conducting further analyses of Burgers vectors and impurities in nanopipes to identify the other factor.

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**Appendix**

C–V results for LP-PNDs, MP-PNDs, and AP-PNDs are shown in Fig. (A·1). They show good linearity, indicating that the doping is uniform with the depth. SIMS data for LP-PNDs and AP-PNDs are shown in Fig. (A·2). The concentration of Si was controlled to about $2 \times 10^{16} \text{cm}^{-3}$ in the drift layer and that of Mg was controlled to $4 \times 10^{19} \text{cm}^{-3}$ in the p-GaN layer. The amount of O was under the detection limit. The concentration of C was approximately $7 \times 10^{15} \text{cm}^{-3}$.

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**Fig. (A·1).** (Color online) C–V characteristics of LP-PNDs, MP-PNDs, and AP-PNDs with electrode diameter of 340 μm.

**Fig. (A·2).** (Color online) SIMS profiles of LP-PNDs and AP-PNDs after annealing. The detection limits for Mg, Si, C, and O were $1 \times 10^{15}$, $7 \times 10^{14}$, $4 \times 10^{15}$, and $6 \times 10^{15} \text{cm}^{-3}$, respectively.
