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Influence of Inversion Domains on Formation of V-Shaped Pits in GaN Films

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The influence of inversion domains (IDs) on the formation of V-shaped pits in GaN films grown by metalorganic chemical vapor deposition has been investigated using transmission electron microscopy. It was found that IDs can induce V-shaped pits which have a large size distribution. Upon introducing InGaN/GaN multiple-quantum wells to observe various growth stages, our results showed that the origin of these ID-induced pits may be a delayed formation of the IDs during island-island coalescence at the initial stage of film growth, thus we need not adopt a general explanation based on the different growth rates for the two opposite polarities.

KEYWORDS: gallium nitride film, inversion domain, V-shaped pit, TEM

Group-III nitride-based alloys have been intensively investigated since the development of GaN-based devices for high-brightness blue/green light-emitting diodes and continuous-wave laser diodes. These GaN-based layers are primarily grown on sapphire substrates which generally contain several kinds of defects such as threading dislocations, stacking faults, voids, inversion domains (IDs), and V-shaped pits.[1)] V-shaped pits are generally associated with threading dislocations, and occur particularly easily in layers containing InGaN[2–5)] or AlGaN[2)] multiple-quantum well (MQW) structures as well as ID sites.[2, 6)] For the latter case, a model based on the difference in growth rates between the N-polar inversion domain and the Ga-polar matrix has been proposed to explain the formation of those ID-induced pits.[2, 6)] Similarly, the appearance of pyramids or hillocks on the sample surface related to IDs has been attributed to a faster growth rate of the Ga-polar domains compared with that of the N-polar matrix.[7–9)] Liliental-Weber *et al*.[2)] have reported that the growth rate in the Ga-polarity region is nearly two times higher than that of a matrix with N-polarity under identical growth conditions. However, in most other reports, either for V-shaped pits or hillocks, the difference in the growth rates of regions of two opposite polarities was estimated to be small.[6–9)] Furthermore, the large size distribution of pits and/or hillocks[6, 7)] within a layer could not be explained based on the different growth rates of regions with opposite polarities. Therefore, it is necessary to clarify the relationship between IDs and V-shaped pits. In this study, we carried out a detailed investigation regarding this issue, using transmission electron microscopy (TEM). We show clear evidence that V-shaped pits of various sizes are formed mainly due to a *growth delay* of the IDs with respect to the surrounding matrix rather than a difference in growth rates.

The sample was grown on a (0001) sapphire substrate by atmospheric-pressure metalorganic chemical vapor deposition (MOCVD). The substrate was initially treated in H_2 ambient at 1150°C, followed by the growth of a 25-nm-thick low-temperature (450°C) GaN buffer layer and a thick undoped GaN layer with a total thickness of 2.6 μm grown at a high temperature of 1075°C. In this sample, two identical InGaN/GaN MQW structures are introduced: one is 0.6 μm and the other is 1.3 μm from the sapphire substrate. The MQWs were grown at a temperature of 700°C, and consisted of ten periods of 6-nm-thick GaN barriers and 4-nm-thick $In_{0.1}Ga_{0.9}N$ wells. The cross-sectional TEM sample was prepared by a standard mechanical polishing and ion-thinning technique to achieve electron transparency along the [11−20] zone axis. In addition, plan-view specimens were produced by thinning the samples from the back. TEM observations were carried out using a Philips CM200 TEM system operated at 200 kV.

Figure 1 shows the low-magnification cross-sectional TEM

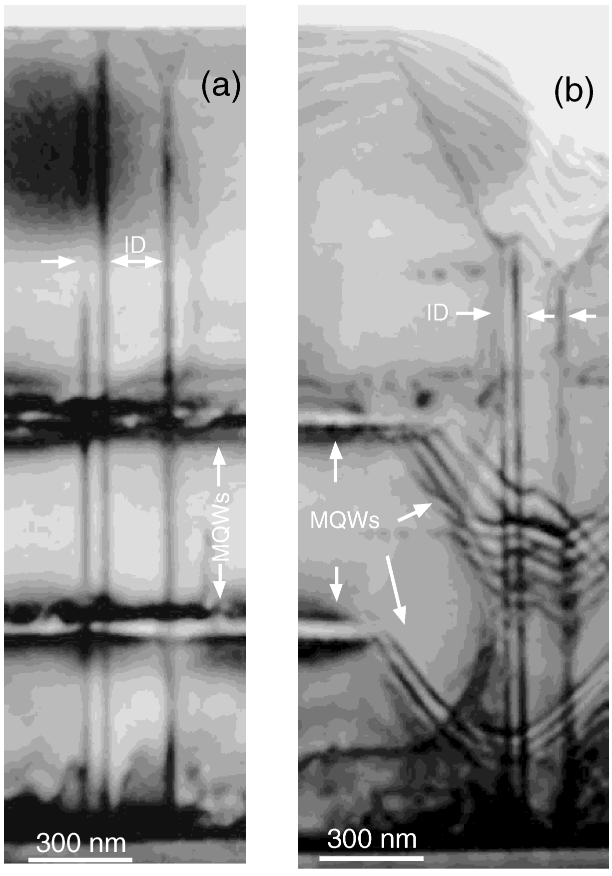

Fig. 1. TEM cross-sectional image of the sample with InGaN/GaN multiple quantum wells (MQWs) having (a) small and (b) large V-shaped pits related to inversion domains. The dark contrasts in the middle of the film parallel to the (0001) plane or with V-shape are MQW layers.

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image of the sample with InGaN/GaN MQWs taken near the [11−20] zone axis with $g = (0002)$, in which some V-shaped pits connected to columnar IDs at their bottom apex clearly appear. (The IDs can be identified by means of high-resolution TEM (HRTEM) and multiple-beam dark-field images taken with $g = \pm(0002)$ along the non-centrosymmetric [11−20] zone axis.[10)]). The pits generally have inclined walls faceted on {1−101} planes. Figure 2 shows a HRTEM image of a V-shaped pit sidewall. The V-shapes have an angle of about 57° between the arms, which is very close to the calculated angle (56.1°) between two polar {1−101} planes. This means that the structural features of these V-shaped pits agree well with those in previous reports.[2,6)] In this sample, almost all pits are connected to columnar IDs, while most threading dislocations do not lead to the formation of V-shaped pits, as shown in Fig. 3. Furthermore, the density of the V-shaped pits is estimated to be about $1 \times 10^7/\mathrm{cm}^2$ from the plan-view TEM images.

The majority of the V-shaped pits are between 100–200 nm in diameter, as shown in Fig. 1(a), while a few pits are 500–800 nm and even 1–1.5 μm in diameter, such as those in Fig. 1(b) and plan-view images (not shown). The model of the formation of V-shaped pits due to the different growth rates between different polarities[2,6–9)] cannot explain the large size distribution of ID-induced pits within the same sample. In the discussion below, we demonstrate that a delay in the growth of IDs with respect to the matrix plays a more important role in the formation of these pits than the difference in the growth rates.

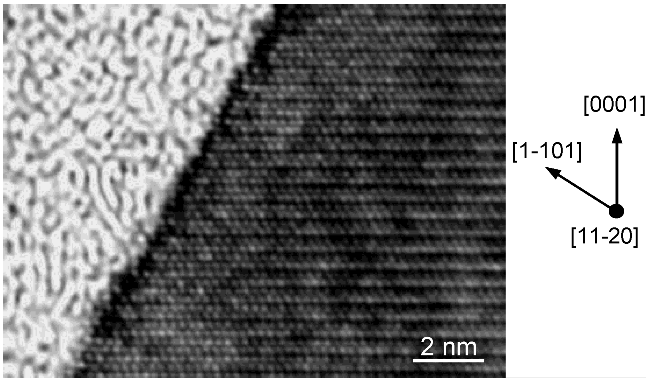

Fig. 2. High-resolution TEM image of one sidewall of a V-shaped pit.

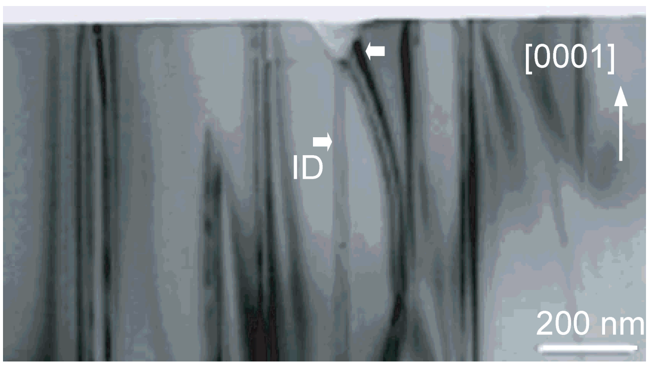

Fig. 3. A V-shaped pit connects to an inversion domain while no pit originates from threading dislocations. Note that a dislocation bends and terminates on an inclined facet of the ID-induced V-shaped pit. Since dislocations cannot terminate within a crystal; this means that the V-shaped pit is empty.

Since there is a clear contrast between InGaN and GaN layers due to a difference in atomic mass density, we introduced an InGaN/GaN MQW structure as a monitor to observe the film structure at various growth stages. If there is an open hexagonal inverted pyramidal pit during growth, the subsequent InGaN layer will be deposited on the sidewall of the pit. Therefore, one can see a V-shape in the TEM image due to the contrast between GaN and InGaN layers,[4)] as shown in Fig. 1(b). Thus, the observation of V-shapes appearing near the substrate in Fig. 1 could be attributed to the deposition of InGaN/GaN layers on the inclined sidewalls of a V-shaped pit, which means that the V-shaped pit formed prior to the introduction of the InGaN/GaN layers. (It also implies that the V-shaped pits are not induced by the introduction of InGaN/GaN layers.) In our sample, the V-shaped pits due to IDs propagate through the entire film keeping their original features, as seen from the two MQW contrasts related to the same ID having a similar V-shape.

Generally, after high-temperature annealing of the low-temperature GaN buffer, the growth of GaN starts from isolated islands which have pyramidal walls of {1−101} planes and a (0001) top plane; then island coalescence occurs, which leads to a V-shaped junction region.[11)] Therefore, one can assume that the V-shape near the substrate, as shown in Fig. 1(b), results from the coalescence of surrounding islands. Also, it is known that the growth of GaN (including IDs) in the junction region could be delayed compared with the as-grown surrounding islands. Generally, the subsequent growth in the junction region should be faster than that of the (0001) plane, resulting in a smoothing process that produces a flat (0001) top surface.[11)] However, the above results suggest that, due to the existence of an ID within the island-island junction region, the conventional smoothing process stops and the V-shaped pit formed can extend to the top surface of the film. The delayed ID grown in association with the coalesced islands plays a key role in the formation of the V-shaped pits, which is different from the previous explanation[2,6)] in which V-shaped pits result from a difference in the growth rates between the ID and matrix. If so, the large size distribution of the V-shaped pits can be explained simply by the variations in the initial thickness of the surrounding islands coalescing with IDs.

In order to illustrate the process clearly, we give a schematic illustration in Fig. 4 based on the TEM image, as shown in Fig. 1. The pit labeled “A” in Fig. 4 indicates that nucleation island coalescence is delayed with respect to the coalescence of the pit labeled “B”, i.e., when the islands coalesce and an ID is formed at the B-pit (dotted line), the coalescence of islands around the A-pit (dotted line) does not occur. Consequently, the small islands coalesce to large thickness and form a V-shaped pit. Therefore, the thickness of the matrix at the coalescence moment should then determine the ID-related pit size, at least initially.

The lines labeled MQWs in Fig. 4 correspond to the InGaN/GaN MQW contrasts in Fig. 1 and are used to identify the *simultaneous* growth in different regions. The correspondence between the A-pit in Fig. 4 and the TEM image in Fig. 1(b) yields useful information. By measuring the distance between the two MQWs, the growth thicknesses of the ID

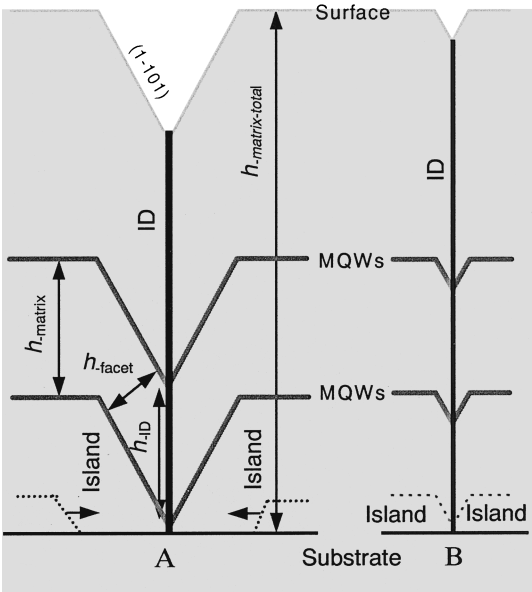

Fig. 4. Schematic illustration of inversion domains (IDs) leading to the formation of V-shaped pits with different sizes. The picture is drawn based on the TEM image, as shown in Fig. 1. The formation of the A-pit is delayed with respect to that of the smaller pit labeled B. The dotted line represents the surface at the time when the B-pit was formed, while the A-pit was not formed.

($h_{\mathrm{ID}} \approx 0.6\,\mu$m) and the matrix (*h-matrix* $\approx 0.6\,\mu$m), as well as of the inclined facet ($h_{\mathrm{facet}} \approx 0.3\,\mu$m), can be obtained, which provides information on the various growth rates. The growth rates of the matrix and the ID are almost equal. This further confirms that the pit sizes are mainly determined by their initial size at the moment of coalescence, but not the different growth rates between the matrix and IDs. Furthermore, the growth rate of the $\{1-101\}$ planes decreases by a factor of 2 compared with that of the IDs (and the matrix), which is in good agreement with the result of crystallographic analysis that the growth rate ratio of ID and $\{1-101\}$ planes should be about 2 (i.e., $1/\sin(56.1^\circ/2)$) to maintain the calculated angle of 56.1° between the two $\{1-101\}$ planes of the V-shaped pit during the growth process.

Based on the above discussion, when an ID forms at a junction of the coalesced islands during the initial growth stages, the ID-induced V-shaped pits will have different sizes according to the thickness of the as-grown surrounding islands. In fact, some IDs could also originate within an isolated island and grow simultaneously with the matrix, in which case no delayed growth of the ID occurs with respect to the matrix. In this case, the ID-induced pit sizes could depend on other growth parameters such as the slightly different growth rates between the two polarities and growth steps from the substrate or buffer layer. Therefore, one can predict that the pits have small size, and indeed, some V-shaped pits related to IDs are observed, in the same sample, to be of very small size (or depth) (< 50 nm) in comparison to those shown in Fig. 1. Finally, from the point of view of crystal growth, since nearly all IDs originate from the buffer layer and/or substrate, we believe that by changing the surface status of sapphire substrates and the initial growth conditions of buffer layers, the number of IDs and ID-related V-shaped pits may be decreased.

In conclusion, we have discussed the formation and growth mechanism of V-shaped pits related to IDs with a large size distribution. We estimated that the relative growth rates between the IDs and the matrix should be almost equal, and then proposed a model to explain the formation of V-shaped pits based on a delay in the growth of the IDs with respect to the matrix. The initial growth stage therefore plays an important role in determining the properties of ID-induced V-shaped pits. This information suggests that it is necessary to suppress the formation of IDs during the initial growth to improve the quality of GaN-based epitaxial films.

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