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Growth Mechanism of Nonpolar A-Plane GaN on Patterned M-Plane Sapphire

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1. Introduction

Plenty of heterosubstrates have been adopted by research communities for the epitaxy of nonpolar or semipolar GaN, such as $LiAlO_2$, *r*-plane sapphire, and *m*-plane sapphire [1-3]. These results are promising, but still numerous myriads of obstacles to overcome to realize further advances in GaN optoelectronics. One of the most pressing challenges is lowering the density-level of extended structural defects in nonpolar or semipolar GaN. Fortunately, these notorious extended defects can be reduced by employing sidewall epitaxial lateral overgrowth (SELO) [4]. Nevertheless, sapphire is still the most common substrate for epitaxial GaN due to its relatively low cost, high temperature stability, and large diameter. Ni *et al.* and Baker *et al.* reported the growth of $(10\bar{1}\bar{3})$ and $(11\bar{2}2)$ GaN films on *m*-plane sapphire [5, 6]; Okada *et al.* demonstrated the fabrication of *m*-plane GaN films by direct epitaxial lateral overgrowth from *c*-plane sapphire sidewalls (ELOSS) [7-9]. However, the cause of selective area epitaxy (SAE) of trench-patterned *m*-plane sapphire without SiO_2 mask is still unclear.

This study demonstrated nonpolar *a*-plane GaN layers on trench-patterned *m*-plane sapphire. The GaN layers were selective deposited on etched *c*-plane sidewalls, rather than *m*-plane terraces. Furthermore, the SAE of *c*-plane GaN on *c*-plane sidewalls and $(11\bar{2}2)$ GaN on *m*-plane terraces were investigated via tuning the V/III ratio.

2. Experiments

The trench-patterned nonpolar *m*-plane sapphire substrate was fabricated by e-gun evaporated nickel mask and Cl_2-based inductively coupled plasma reactive ion etching (ICP-RIE). Symmetric crystallographic *c*-plane facets of sapphire with terrace/trench width 4μm/2μm and 3μm etching depth were illustrated in Fig. 1. To effectively expose the *c*-plane sidewalls, terrace/trench stripes orientation were chosen parallel to $[11\bar{2}0]_{sapphire}$. The growth of GaN on the trench-patterned substrate was performed on by Metal Organic Chemical Vapor Deposition (MOCVD) system. A 30 nm GaN buffer layer was grown at 550°C, followed by main GaN growth at 1050°C with a pressure of 100 mtorr. The growth mode of nonpolar GaN on trench-patterned *m*-plane sapphire was achieved via tuning the V/III ratio from 72 to 350, 1800 and up to 9000. Additionally, an optimized substrate with lower terrace/trench width ratio (*i.e.* 2um/4um) is used to attempt to suppress the growth of GaN from the $(10\bar{1}0)_{sapphire}$ terraces under the V/III ratio of 72. The grown samples were characterized by ω- and 2θ-scans of X-ray diffraction (XRD), scanning electron microscopy (SEM) and transmission electron microscopy (TEM).

3. Results and discussion

The exposed symmetric *c*-plane sidewalls and *m*-plane terraces of sapphire during MOCVD growth resulted in two kinds of GaN crystallites; each GaN crystallite has unique growth orientation and terminal planes. First, on-axis 2θ-ω XRD scans (not shown here) indicated that pure *a*-plane GaN was carried out under the V/III ratio of 9000, 1800, and 350. Fig. 2(a) ~2(c) showed that the SAE of *c*-plane GaN was grown laterally only from the etched *c*-plane sidewalls. On the contrary, deposition of GaN was completely absent on the *m*-plane terraces. It's confirmed that the crystallographic orientation relationships of GaN stripes on patterned *m*-plane sapphire are $(11\bar{2}0)_{GaN} \parallel (10\bar{1}0)_{sapphire}$, $[0001]_{GaN} \parallel [0001]_{sapphire}$, and $[10\bar{1}0]_{GaN} \parallel [11\bar{2}0]_{sapphire}$ [5].

The SEM images in Fig. 3 show clearly the representative cross-sectional morphology of GaN stripes selectively grown on trench-patterned *m*-plane sapphire. The shapes of GaN stripes were a strong function of the V/III ratio. Fig. 3(a) shows the sample carried out with a V/III ratio of 9000 for 30 min. GaN grew from the *c*-plane trench sidewalls only with faster growth rate along +*c* direction than that in *a* direction, and therefore the upward planes are inclined by 58.5° with respect to the trench sidewalls. Additionally, the upwardly inclined plane was tentatively attributed to $(11\bar{2}2)$ planes, since the growth front was constructed by the competition between +*c* planes and *a* planes of GaN.

Fig. 3(b) revealed that when the V/III ratio was decreased to 1800, the growth mode is similar to that with a V/III ratio 9000 for 30 min. The reduction of V/III ratio led to a more conspicuous increase of growth rate in both +*c* and *a* direction of GaN which leads to the coalescent tip and level-raising respectively. As a matter of fact, the upwardly inclined $(11\bar{2}2)$ planes are maintained.

Next, a different growth mode was presented under a still lower V/III ratio, *i.e.* 350, with the same growth time. As in Fig. 3(c), the growth along the +*c* direction advanced overwhelmingly faster than that in *a* direction and was stymied by itself while these planes coalescence in +*c* direction. This formed the upwardly flat *a*-plane. Something must be mentioned that the growth along the -*c* direction was almost stationary, no matter how much the V/III ratio vary, from 9000 to 350. Furthermore, the growth on the $(10\bar{1}0)$ sapphire terraces has been completely forbidden.

Fig. 2(d) showed the the dramatically-altered growth mode and crystal orientations by decreasing the value of

V/III ratio to 72. The GaN grown laterally (from *c*-plane sidewalls) and vertically (from *m*-plane terraces) were obtained simultaneously which revealed in Fig. 3(d). The GaN stripes grown from *c*-plane sidewalls performed similar behavior with that at V/III ratio of 350, but it played another role as the mask for the epitaxial lateral overgrowth (ELO) of GaN on the *m*-plane terraces. These well-defined in-situ-forming GaN-masks in $[11\bar{2}0]_{sapphire}$ direction suppressing the growth of $(10\bar{1}1)$ GaN on *m*-plane terraces and leading to pure growth of $(11\bar{2}2)$ GaN, and the in-plane epitaxial relationships of $(11\bar{2}2)$ GaN on *m*-plane terraces are $(11\bar{2}2)_{GaN} \parallel (10\bar{1}0)_{sapphire}$, $[10\bar{1}0]_{GaN} \parallel [\bar{1}2\bar{1}0]_{sapphire}$, and $[\bar{1}2\bar{1}\bar{1}]_{GaN} \parallel [000\bar{1}]_{sapphire}$ [5]. It's worth noting that the gradual growth of lateral-GaN limited the ELO of $(11\bar{2}2)$ GaN. As a result, the $(11\bar{2}2)$ GaN shows no noticeable wings over the lateral-GaN but it contains 32° inclined +*c* planes with respect to the upwardly flat $(11\bar{2}2)$ plane.

Fig. 4 shows the TEM image of GaN under a V/III ratio of 72 on a lower terrace/trench width ratio (2μm/4μm) substrate. The growth mode is equivalent to that of higher terrace/trench width ratio (*i.e.* 4μm/2μm); meanwhile, the SFs were localized in the GaN grown from the *m*-plane terraces. The orientation of SFs in the GaN on the *m*-plane terraces, completely parallel to the inclined (0001) GaN planes, are in close correlation with its growth rate along the +*c* direction, which advances overwhelmingly faster than that in *a*-direction. Apparently, the narrower terraces did effectively restrain the volume of semipolar $(11\bar{2}2)$ GaN growing from *m*-plane terraces; however, the growth of GaN cannot be eliminated by decreasing width of *m*-plane terraces. In short, the growth mode is strongly depends on the V/III ratio during MOCVD growth rather than the terrace/trench width ratio of the patterned substrate.

4. Conclusions

In this paper the growth mechanism of *a*-plane GaN on trench-patterned *m*-plane sapphire by MOCVD under different V/III ratio was demonstrated. For V/III ratio from 350 up to 9000, the lateral growth of GaN from the +*c* plane sidewalls is dominated. Under low V/III ratio of 72, the growth of lateral-GaN accompanied the aroused $(11\bar{2}2)$ GaN on *m*-plane terraces. This unexpected appearance of semipoalr $(11\bar{2}2)$ GaN can be suppressed by optimizing the V/III ratio. On the other hand, lower terrace/trench width ratio doesn't interdict it. By optimizing V/III ratio and appropriate terrace/trench width ratio of patterned *m*-plane sapphire substrate, we expect that a high quality nonpolar *a*-plane GaN on *m*-plane sapphire is achievable.

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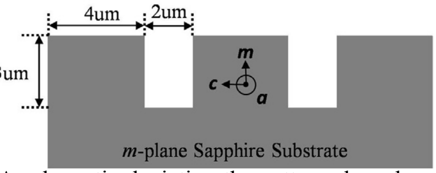

Fig. 1 A schematic depicting the patterned *m*-plane sapphire substrate with 4μm terrace width, 2μm trench width and 3μm depth.

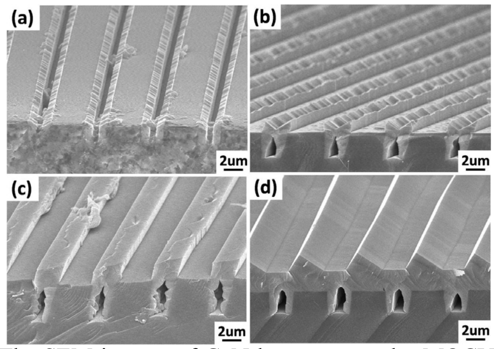

Fig. 2 The SEM images of GaN layers grown by MOCVD under the V/III ratio of (a) 9000, (b) 1800, (c) 350 and (d) 72.

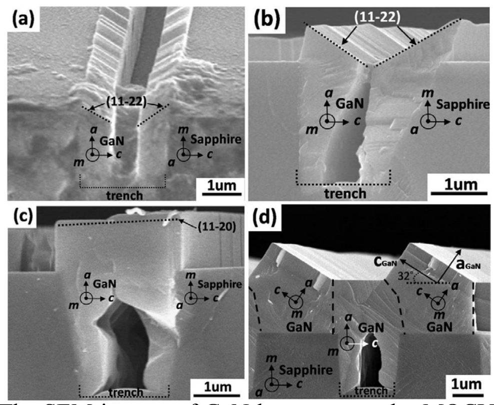

Fig. 3 The SEM images of GaN layers grown by MOCVD under the V/III ratio of (a) 9000, (b) 1800, (c) 350 and (d) 72. The corresponding crystal planes and orientations are depicted.

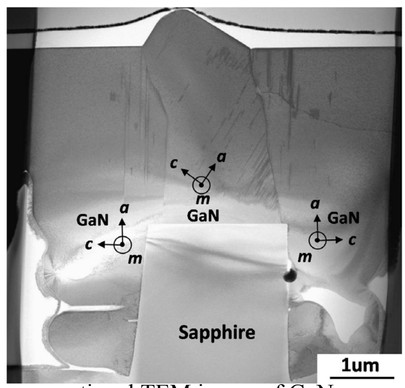

Fig. 4 The cross-sectional TEM image of GaN grown by MOCVD under the V/III ratio of 72 with terrace/trench width 2μm/4μm. The corresponding crystal planes and orientations are depicted.