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Observations on surface morphologies and dislocations of a-plane GaN grown by metal organic chemical vapor deposition

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In this study, we grew non-polar a-plane GaN thin films on r-plane sapphire using a series of growth conditions by metal-organic chemical vapor deposition. The results showed that high temperature and low-pressure conditions benefited two-dimension growth could lead to a fully coalesced a-plane GaN layer with a very smooth surface. The best surface morphology with an excellent mean roughness of 10.5 Å was obtained. The different thickness AlN as a nucleation layer and the different δ/β ratio were also considered. The results revealed that the surface morphology would get worse when the thickness of nucleation layer and δ/β ratio were away from the values of optimal condition. The observation of transmission electronic microscopy shown the lowest density of threading dislocations was 1.85×10^{10} /cm^2.

1 Introduction

GaN-based semiconductors and their heterostructures have recently attracted considerable interest due to their potential for device applications, such as visible and ultraviolet light-emitting diodes (LEDs) [1], laser diodes (LDs) [2] and high–power transistors [3] deposited on either basal plane (0001) sapphire, SiC, or free standing GaN substrates. Unfortunately, epitaxy toward the (0001) orientation leads to undesirable spontaneous and piezoelectric polarization effects, resulting in quantum confined Stark effect (QCSE), which would significantly reduce the carrier recombination rate from quantum wells grown on polar substrate orientations [4, 5]. To eliminate such polarization effects, growth along non-polar orientations has been explored for a-plane GaN on r-plane sapphire and a-plane SiC, and m-plane GaN on [100] $LiAlO_2$ substrates. [6, 7]

Growth of non-polar group-III nitride hexagonal heterostructures could overcome the presence of large built-in electrostatic fields and also could improve the quantum efficiency of light emitting diodes. However, the surface morphology of the GaN grown on r-plane sapphire was usually quite rough, making device fabrication more difficult. As a result, most groups avoided using r-plane sapphire substrates to grow GaN devices. In order to overcome the influence of defects generated due to the lattice mismatch, Chitnis et al. grown a-plane LED structure on r-plane sapphire by using a GaN with a thickness of more than 30 μm [8]. Haskell et al. presented an approach to employ epitaxial lateral overgrowth (ELOG) [9]. Although ELOG method could yield a-plane GaN films over r-plane sapphire with significantly improved surface morphologies and with reduced dislocation densities, this approach gets complicated and consumed much time.

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In this study, the optimal growth condition of the a-plane GaN layers with pits-free and smooth surface were investigated in detail by modifying growth temperatures, pressures, and V/III ratio. Furthermore, the influences of different thickness of AlN as nucleation layers were also considered. Eventually, we obtained a high quality a-plane GaN bulk with a smooth surface that is capable of fabricating other device structures, such as multiple quantum wells and LEDs.

2 Experiments The r-plane sapphire substrates were first annealed at 1400 °C in a thermal furnace for 1 hour. After chemical cleaning, sapphire substrates were loaded into the low-pressure metal-organic chemical vapor deposition (MOCVD) system. We used low-temperature AlN as nucleation layers instead of traditional GaN layers. The nucleation layer growth temperature and thickness were 600 °C and 30 nm, respectively. In order to further optimize the growth conditions for a-plane GaN films, influence of different pressures (100, 200, and 300 Torr) and temperatures (1020, 1070 and 1120 °C) on growth of 2 μm thick GaN bulks was studied. Trimethylgallium (TMGa), trimethylaluminum (TMAl) and ammonia were used as Ga, Al and N sources, respectively. Then, we fixed the growth pressure and temperature, which would result in the smoothest surface morphology, and modulated different thickness of AlN nucleation layers from 30 nm to 15 and 45 nm. In addition, the V/III ratio for growth of the GaN bulk was adjusted from 900 to 600 and 1200, subsequently. The surface morphologies of all samples were characterized by optical microscopy (OM) with x magnification, scanning electronic microscopy (SEM) and atomic force microscopy (AFM). The distribution of dislocations was observed by transmission electronic microscropy (TEM).

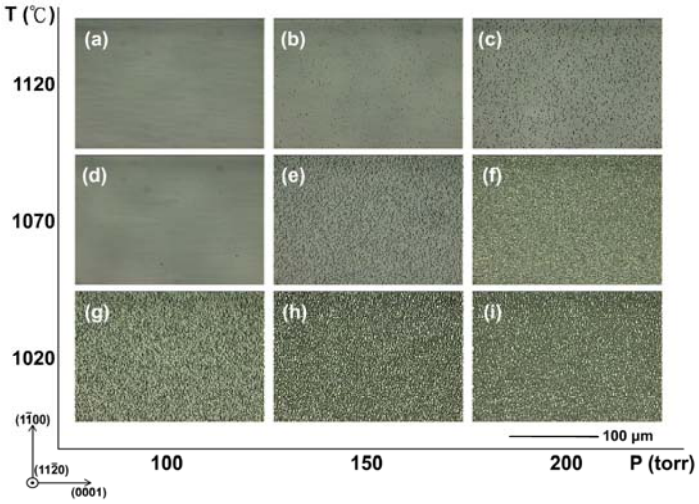

Fig. 1 OM images of a-plane GaN surfaces for growth temperatures of 1020–1120°C and pressures of 100–300 Torr.

3 Results and discussion For the first step, we'd like to study the optimal growth parameters in growth pressure and temperature. We fixed the thickness of AlN nucleation layer as 30 nm and the V/III ratio for growing a-plane GaN bulk as 900. In order to make all analysis easy to understand, we set our results and growth parameters into a three by three square chart. Figure 1 shows the typical OM images of a-plane GaN with different growth conditions. We found the nucleation island did not coalesce fully causing the surface rugged and rough when the growth pressures were high and temperatures were low, such as shown in Figs. 1(f), (h) and (i). Once the growth temperature was elevated and pressure was decreased as shown in Figs. 1(b)–1(e) and 1(g), the nucleation islands gradually coalesced with each other and remained pits distributed on the sample surface. Figure 1(a) manifested smoothest surface morphology without any observable pits when the growth temperature and pressure were set as 1120 °C and 100 Torr, respectively.

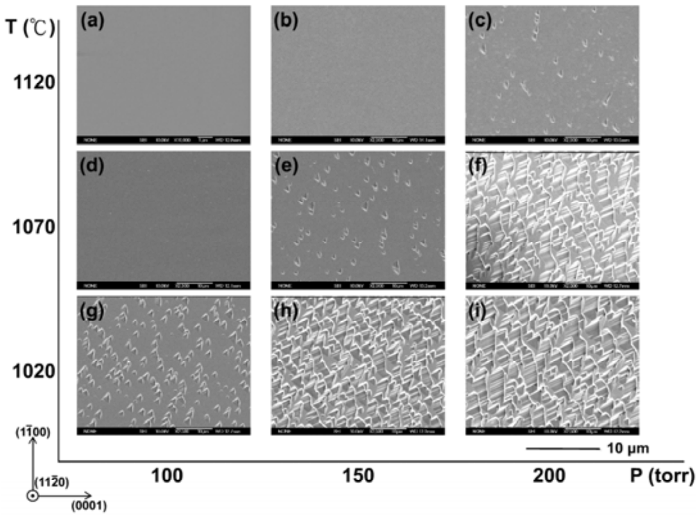

Fig. 2 SEM images of a-plane GaN surfaces for growth temperatures of 1020-1120 °C and pressures of 100–300 Torr.

We enlarged the images of surface morphology taken by SEM and showed the results as in Fig. 2. The growth condition of high temperature and low pressure was shown in Fig. 2(a) still revealing the best surface morphology in terms of flatness. However, we could clearly observe not only many pits existed but also wavy stripes formed along c-axis in Figs. 2(f), (h) and (i). The possible reason of strip formed was due to the overgrowth by fast growing along c-direction. On the other hand, except for triangle pits, the irregular strips could not be observed in Fig. 2(b)–(e) and (g). In general, the common feature of V defects is characteristically observed when c-plane GaN grown in the kinetically limited condition [10]. The V defect for a c-plane GaN is an inverted pyramid bound by the pyramidal {10-11} facets. Since the V defect only appears it's half side in an a-plane GaN under the non-polar growth scheme, such kind of pits was named a < defect originating from upon island growth and coalescing at the initial growth of high temperature GaN bulk [11]. Above the all analysis, we could suggest that the growth mechanism of a-plane GaN at the beginning stage followed a Volmer-Weber (VW) mode which would lead to the three dimension (3D) growth. Each GaN column was probably grown from a GaN island formed on top of AlN nucleation layer. After most of the islands merged to each other, a Frank-van der Merwe (F-vdM) mode occurred and dominated the subsequent growth process, which would lead to two dimension (2D) growth. The 2D growth condition is necessary for thick a-plane GaN since the difference of growth rate

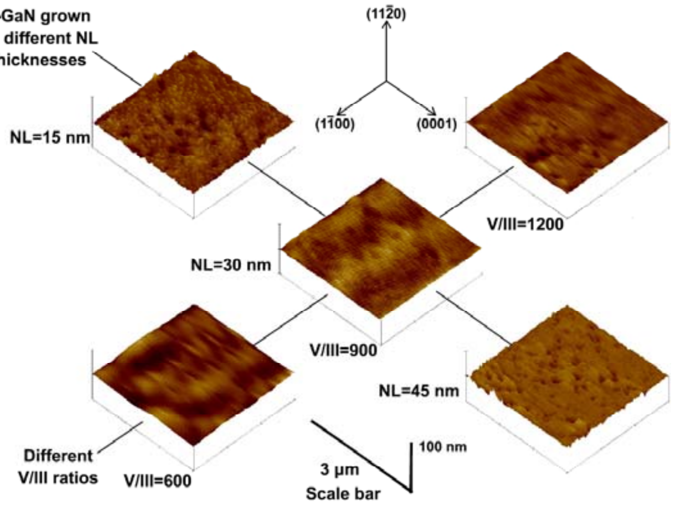

Fig. 3 AFM images of a-plane GaN surfaces for growth of V/III ratio of 600–1200 and on different thickness of nucleation layer.

could occur between c-axis and m-axis. Otherwise, the 3D growth would result in the appearances of < defects and wavy strips.

In addition to the effect of epitaxial temperature and pressure, the thickness of the nucleation layer and the V/III ratio for growth of the a-plane GaN bulk were also considered. The growth pressure and temperature were fixed to the same values as those resulting in the best surface flatness and the thickness of the nucleation layer and the V/III ratio were modulated. The thickness of nucleation layer was adjusted from 30 nm to 15 and 45 nm. And the V/III ratio was adjusted from 900 to 600 and 1200. The surface morphologies of all samples were observed by AFM and the results were shown in Fig. 3. The center 3D AFM image is the result of the sample grown at 1120 °C and at 100 Torr, which shown submicro pits and additional stripe features along the [0001] direction. Meanwhile, the sample showed very smooth surface with a root mean square (RMS) roughness of only 10.5 Å. Such roughness was very far less than the previous report by Ni et al., whose sample was grown at lower growth temperature on a thicker nucleation layer [12]. However, the tiny surface morphologies would get worse when we used thinner and thicker AlN nucleation layer as template, beside there were many pits with size around 100 nm. Owing to that each GaN column is probably grown from a GaN nucleus formed on top of each columnar AlN region. The columns of AlN have suitable disordered orientations for GaN. Once the growth condition has been changed, it would cause the variation of disordered orientations. In our growth condition, therefore, we suggested the 30 nm of AlN is the optimal thickness for growth of a-plane GaN due to the suitable disordered orientations of nucleation layer.

On the other hand, the surface morphology got worse when the V/III ratio was increased or decreased from 900. The high V/III ratio leaded to high NH3 flow injected and further produced more amounts of Hydrogen during the growth process. Some pits distributed on the surface under the high V/III ratio could be due to the excess Hydrogen facilitating the surface dissociation rate and damaging the surface morphology [13]. Although the low V/III ratio condition benefits the 2D growth, some Ga atoms would not react with insufficient N atoms causing a wavy surface. To sum up above discussions, we suggested both the thickness of nucleation layer and the V/III ratio have a optimal value to be suitable to grow a-plane GaN bulk.

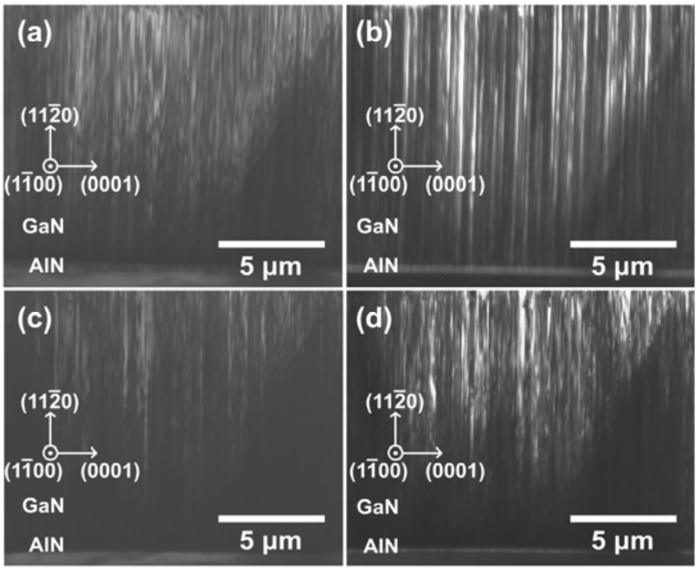

Fig. 4 Bright and dark field cross-section TEM images of GaN epilayer grown under optimal condition with g vector [11-20] for (a),(b) and [0002] for (c),(d).

In order to realize the extended defects in the GaN epilayer, cross-sectional transmission electron microscopy was employed. Figure 4 shows cross-sectional TEM image along [1-100] zone axis of the GaN epilayer grown under optimal condition, and two beam diffraction contrast bright and dark field images with g vector [11-20] (as shown in Figs. 4(a-b)) and [0002] (as shown in Figs. 4(c-d)) were shown, respectively. According g*b dark field analysis, most of threading dislocations in the GaN films in contrast in both of g = [11-20] and [0002] dark field images and hence should be mixed a+c type dislocations. In our TEM observation, we could calculated the dislocation density to be around 1.85×10^{10} /cm^2. Remarkably, although the high dislocation density in the GaN can be observed in our case, the surface of the GaN epilayer was still very smooth. Thus, the threading dislocation might not be directly responsible to rough or pit surface of a-plane GaN grown under other non-optimal condition.

4 Conclusions In conclusion, we grew a-plane GaN with a series of growth conditions included different growth temperatures and the chamber pressures by MOCVD. The results showed that high quality and smooth surface a-plane GaN at high temperature and low pressure growth condition was obtained. In the worse cases of growth, the possible reasons for striped features and surface undulation observed in a-plane GaN were due to the difference of migration lengths of adatoms between c- and m-directions. The best surface morphology with an excellent mean roughness of 10.5 Å was obtained. Moreover, the thickness of the nucleation layer and V/III ratio for GaN bulk growth existed optimal values for smooth surface a-plane GaN growth. In our TEM observation, we could calculated the dislocation density to be around 1.85×10^{10} /cm^2. The result of smooth surface confirmed the feasibility of fabricating non-polar devices on a-plane GaN grown in such growth condition.

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