Defects in GaN Films Grown on Si(111) Substrates by Metal-Organic Chemical Vapour Deposition *

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We report the transmission–electron microscopy study of the defects in wurtzitic GaN films grown on Si(111) substrates with AlN buffer layers by the metal-organic chemical vapour deposition method. The $In_{0.1}Ga_{0.9}N$/GaN multiple quantum well (MQW) reduced the dislocation density by obstructing the mixed and screw dislocations passing through the MQW. No evident reduction of the edge dislocations density by the MQW was observed. It was found that dislocations with screw component can be located at the boundaries of sub-grains slightly in-plane misoriented.

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GaN-based Ⅲ-Ⅴ compounds are the key materials for the fabrication of optoelectronic devices[1−3] in the region of blue-to-ultraviolet. Due to the lack of bulk GaN substrates, GaN is generally grown by hetero-epitaxy. Commonly, threading and misfit dislocations,[4,5] stacking faults[4,6] and inversion domains[6,7] exist in the GaN film. How to reduce the defects density and improve the crystal quality is still of great importance. GaN has been grown mostly on sapphire and SiC substrates. On the other hand, a variety of other substrates, such as Al_2O_3, SiC, Si, GaAs, ZnO, and $LiAlO_2$, have been attempted for the epitaxy of GaN. There have been great interests in growing GaN on Si because Si has high crystal quality, large area size, low manufacturing cost and the potential application in integrated optoelectronic devices. GaN has been successfully deposited on Si substrates[8−13] with AlN, Al_2O_3, silicon nitride, SiC as buffer layers by both metal-organic chemical vapour deposition (MOCVD) and molecular beam epitaxy (MBE) methods. In this Letter, we report the transmission–electron microscopy (TEM) investigation of the defects in the GaN films grown on Si(111) with AlN buffer layers.

Two GaN samples, namely samples A and B, were studied. Both the samples were grown by the horizontal low-pressure MOCVD method. Trimethylgallium (TMGa), trimethylaluminum (TMAl), ethyldimethylindium (EDMIn) and NH_3 were used as the Ga, Al, In and N source materials, respectively. Sample A was grown in the following way. Firstly, about 20 nm AlN was deposited on Si(111). No nitridation was performed before the growth of GaN. Secondly, the epitaxy of about 800 nm GaN at temperature of 1050°C was followed. Thirdly, the five-period nominal $In_{0.1}Ga_{0.9}N$/GaN MQW was grown. Finally, another layer GaN (about 800 nm) was grown at the temperature of 1050°C. For sample B, only about 100 nm GaN layer was deposited on Si(111) with a 20 nm AlN buffer layer.

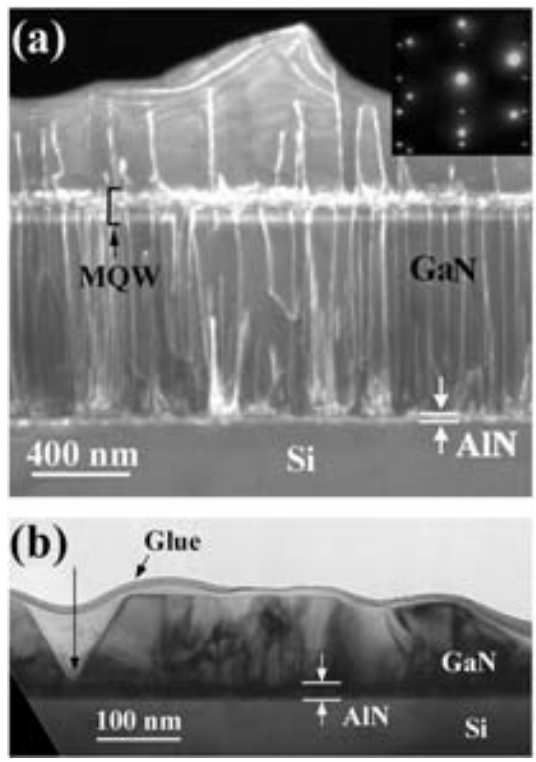


Fig. 1. (a) Dark-field image of sample A. The inset is the composite SAED of the GaN film, the AlN buffer and the Si substrate at the interface region. (b) Bright field image of sample B.

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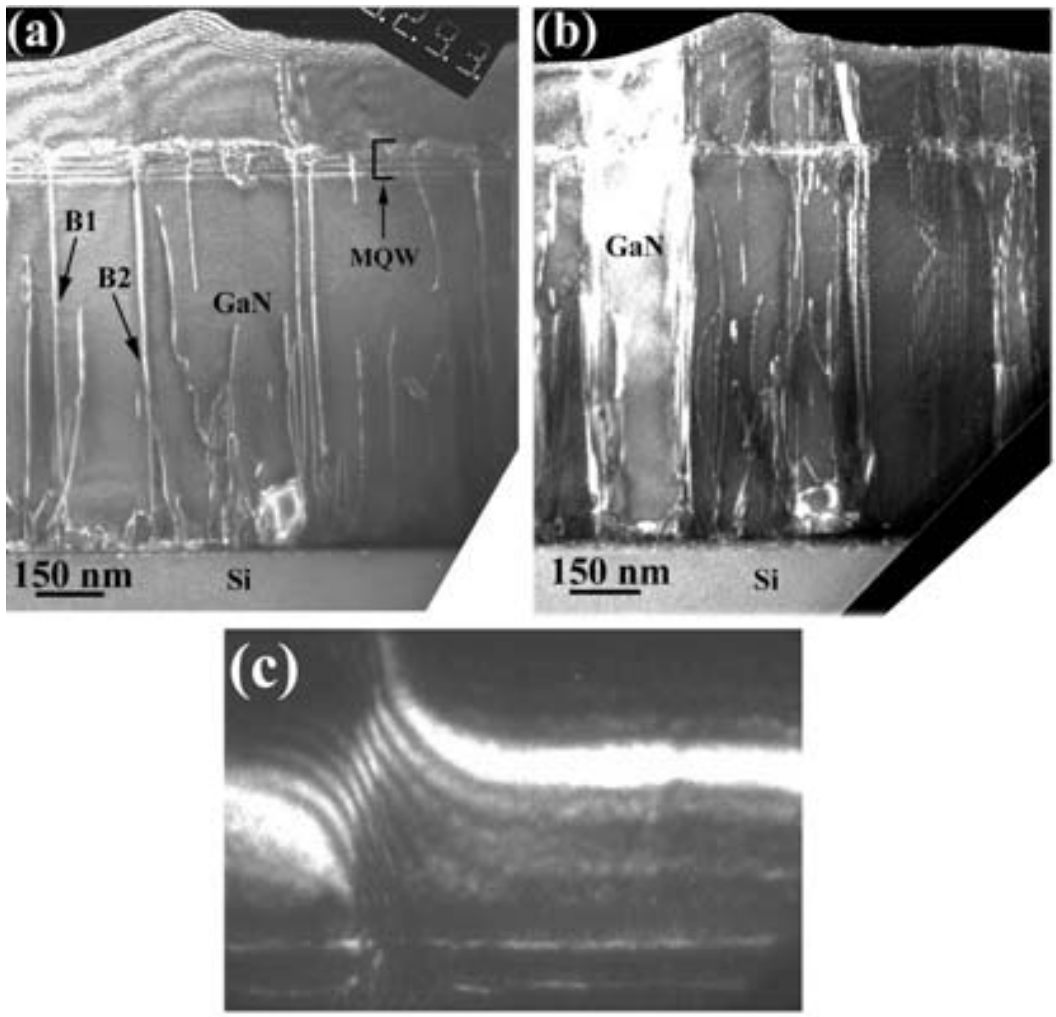


Fig. 2. Transmission–electron micrographs taken from an area of sample A showing the dislocations in the GaN film. (a) Dark-field image with $g = [0002]$ close to the $[11\bar{2}0]$ pole. (b) Dark-field image with $g = [1\bar{1}00]$ close to the $[11\bar{2}0]$ pole. (c) Dark-field LACBED pattern taken from dislocation indicated as B1 in (a). The dislocation diffraction $g = [0008]$ crossing B1 is split into sidebands.

The structure of sample A is shown in Fig. 1(a). The epitaxial relationship was characterized as: $[11\bar{2}0]_{\mathrm{GaN}}//[11\bar{2}0]_{\mathrm{AlN}}//[1\bar{1}0]_{\mathrm{Si}}$, and $(0001)_{\mathrm{GaN}}//(0001)_{\mathrm{AlN}}//(111)_{\mathrm{Si}}$. The critical thickness of GaN grown on Si was calculated as 6.34 nm.[14] Large amount of threading dislocations and half loops exist in the GaN film. The half loops vanish at about 400 nm away from the Si/AlN interface. That indicates that the 3D-2D growth mode transition happened when the GaN layer attained 400 nm. Figure 1(b) shows the structure of sample B. The 3D growth of the GaN can be clearly seen from this image. The sub-grains are still not coalesced.

Commonly, three types of dislocations are present in GaN.[5] They are screw, edge and mixed dislocations with Burger vectors parallel to c, a, and $c + a$, respectively. According to the diffraction contrast theory, the dislocations shown in Fig. 2(a) are mixed and screw threading dislocations and those in Fig. 2(b) are the mixed and edge threading dislocations. Comparison of the two images reveals that the density of dislocations in Fig. 2(b) is much higher than that in Fig. 2(a). That indicates the edge type dislocation is predominant.

When MQW were used, a reduction of threading dislocation density have already been observed.[15,16] This can also be seen from Fig. 1(a). The density of dislocations in the GaN layer beyond the MQW is much reduced. The influence of MQW on different type dislocations and the reduction mechanism are not clear. Figure 2(a) shows that almost all of the dislocations, i.e., screw and mixed dislocations, pass through the lower layers of MQW and stop at the top layers of MQW. Thus the MQW can stop the screw and mixed dislocations and improve the quality of the GaN film. No evident reduction of the density of dislocations can be observed in Fig. 2(b). It can be seen that almost all the edge dislocations pass through the MQW. Edge and mixed dislocations can glide on both the $\{0001\}$ and $\{10\bar{1}0\}$ slip planes. Theoretically, edge and mixed

dislocations can bend either toward another threading line so that they annihilate one another, or normal to the growth direction along the interface so that they do not propagate further into the epilayer beyond the MQW. On the other hand, the screw dislocations can propagate into the upper epilayer because the screw dislocations cannot bend. However, what we have observed is contradictory to the prediction of theory.

To confirm the results obtained by the diffraction contrast technique, dark field large angle convergent beam electron diffraction (LACBED) was used to determine the type of the dislocations. The dislocation indicated by B1 in Fig. 2(a) was analysed by dark field LACBED using $\boldsymbol{g} = [0008]$, as shown in Fig. 2(c). The magnitude of the dislocation is determined by the rule $\boldsymbol{g} \cdot \boldsymbol{b} = n$ where n is the number of subsidiary maxima observed in a dark field LACBED image plus 1. It can be obtained from this picture that $n = 8$. This implies that the dislocation located at B1 has a screw component. The dark-field LACBED analysis of the other dislocations reveals that all the dislocations shown in Fig. 2(a) have the screw component and those in Fig. 2(b) have the edge component. The result of dark field LACBED is consistent with that of diffraction contrast.

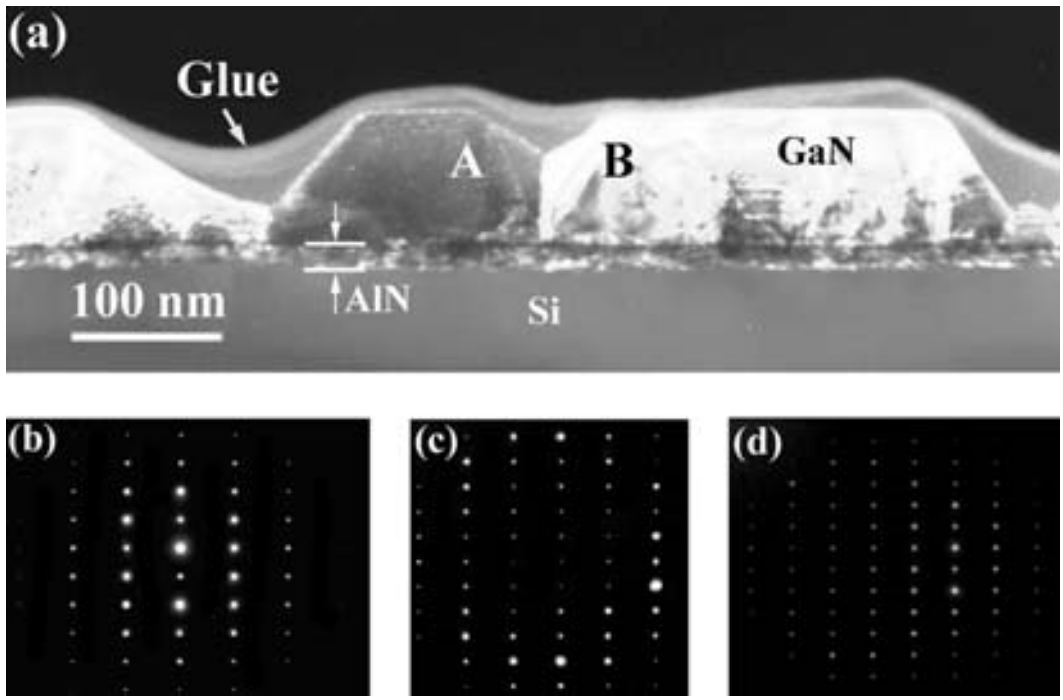


Fig. 3. (a) Dark-field image of sample C taken with $g = [1\bar{1}00]$, showing the mis-oriented sub-grains. (b) SAED of the GaN film in the sub-grain indicated as A in (a). (c) SAED of the GaN film in the sub-grain indicated as B in (a). (d) The combined SAED of sub-grains A and B.

The diffraction contrast and the dark-field LACBED analysis of the dislocations confirm that MQWs can greatly reduce the density of screw and mixed dislocations but cannot effectively reduce the density of edge dislocations. The real mechanism of the reduction of density of dislocations by MQWs need further study.

Bright and dark domain-like contrast can be seen from Fig. 2(b). The domain-like structure shows the sub-grains of the GaN film. The formation of the sub-grains in the GaN film can be seen from Fig. 3(a). Three sub-grains can be observed in this image. Two of them were indicated as A and B sub-grains. Due to the 3D growth at the initial stage of the film epitaxy, the GaN film consists of numerous sub-grains. The contrast in Figs. 2(b) and 3(a) shows that the sub-grains are mis-oriented with each other. Two kinds of mis-orientations, i.e., in-plane twist and out-of-plane tilt, exist in the GaN film. The contrast of the sub-grains in Fig. 2(b) is out of contrast in Fig. 2(a). This indicates that the mis-orientation between the sub-grains is of in-plane twist. The selected area electron diffraction (SAED) patterns of sub-grains A and B, and both the sub-grains were taken respectively, as indicated by Figs. 3(a), 3(b) and 3(c). When the three SAED patterns were taken, the sample was kept at the same X- and Y-tilt angles. The SAED patterns show that the mis-orientation between A and B sub-grains is of in-plane twist and the tilt was determined to be $0.42°$. The SAED analysis of the other sub-grains in the film shows that the mis-orientation is of in-plane twist and the tilt ranges from $0°$ to $1.2°$. It is well known that this kind of the sub-grains is caused by the 3D growth of GaN. When GaN began to nucleate

on AlN, the 3D growth induced the formation of GaN sub-grains. This can be seen form Fig. 1(b). At the left of the image indicated by an arrow, two GaN sub-grains were just formed on the surface of AlN.

The two straight lines B1 and B2 indicated by arrows in Fig. 2(a) are the two boundaries of the sub-grain which shows strong contrast in Fig. 2(b). Threading dislocations located at the boundaries. As mentioned above, the dislocations shown contrast in Fig. 2(a) are screw- or mixed-type dislocations. Thus, the threading dislocations located at boundaries B1 and B2 have screw component. This indicates that dislocations with screw component can be located at the boundaries of sub-grains slightly in-plane mis-oriented. It can be seen from Fig. 2(a) that the threading dislocations with the screw component located at boundaries B1 and B2 are stopped by the MQW. Therefore, there are no dislocations with the screw component located at boundaries B1 and B2 in the upper layer GaN beyond the MQW. This indicates that dislocation with the screw component is not located at every sub-grain boundary. In a previous study,[17] it is reported that pure edge dislocations are located at the boundaries of sub-grains slightly mis-oriented. Thus, it is obvious that the threading dislocations with the edge and screw components all can be located at the boundaries of the sub-grains.

In conclusion, we have studied the defects in hexagonal GaN films grown on Si(111) by the MOCVD method. It was found that the density of mixed and screw dislocations can be reduced by the MQW. No evident reduction of the edge dislocation density by the MQW was observed. The dislocations located at the boundaries of the grains slightly in-plane mis-oriented are also discussed.

References

[1] Nakamura S, Mukai T and Senoh M 1994 *Appl. Phys. Lett.* **64** 1687

[2] Osinsky A, Gangopadhyay S, Yang J W, Gaska R, Kuksenkov, Temkin H, Shimagin I K, Chang Y C, Muth J F and Kolbas R M 1998 *Appl. Phys. Lett.* **72** 551

[3] Sheppard S T, Doverspike K, Prible W L, Allen S T, Palmour J W, Kehias L T and Jenkins T J 1999 *IEEE Electron Device Lett.* **20** 161

[4] Ponce F A, Cherns D, Young W T and Steeds J W 1996 *Appl. Phys. Lett.* **69** 770

[5] Nakajima K, Ujihara T, Miyashita S and Sazaki G 2001 *J. Appl. Phys.* **89** 148

[6] Potin V, Ruterana P, Nouet G, Pond R C and Morkocb H 2000 *Phys. Rev.* B **61** 5587

[7] Wu X H, Brown L M, Kapolnek D, Keller S, Keller B, Den-Baars S P and Speck J S 1996 *J. Appl. Phys.* **80** 3228

[8] Schremer A T, Smart J A, Wang Y, Ambacher O, MacDonald N C and Shealy J R 2000 *Appl. Phys. Lett.* **76** 736

[9] Wang L S, Liu X L, Zan Y D, Wang J, Wang D, Lu D C and Wang Z G 1998 *Appl. Phys. Lett.* **72** 109

[10] Kim M H, Bang Young C, Park N M, Choi C J, Seong T Y and Park S J 2001 *Appl. Phys. Lett.* **78** 2858

[11] Nakada Y, Aksenov I and Okumura H 1998 *Appl. Phys. Lett.* **73** 827

[12] Tanaka S, Kawaguchi Y, Sawaki N, Hibino M and Kazumasa H 2000 *Appl. Phys. Lett.* **76** 2701

[13] Wang D, Hiroyama Y, Tamura M, Ichikawa M and Yoshida S 2000 *Appl. Phys. Lett.* **76** 1683

[14] Hu G Q, Wan L, Kong X, Wang Y Q, Duan X F, Lu Y and Liu X L 2003 *J. Mater. Sci. Lett.* (at press)

[15] Feltin E, Beaumont B, Laü M, Mierry P, Vennéguès P, Lahrèche H, Leroux M and Gibart P 2001 *Appl. Phys. Lett.* **79** 3230

[16] Nikishinm S A, Faleev N N, Antipov V G, Francoeur S, Grave P, Seryogin G A, Temkin H, Prokofyeva T I, Holtz M and Chu S N 1998 *Appl. Phys. Lett.* **75** 2073

[17] Vézian S, Massies J, Semond F, Grandjean N and Vennéguès P 2000 *Phys. Rev.* B **61** 7618