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Characteristics of threading dislocations in ZnO grown on facet-controlled epitaxial overgrown GaN templates

H L Zhou[1,3], S J Chua[2,3], S Y Chow[2], H Pan[1], Y W Zhu[1], Y P Feng[1], L S Wang[2], K Y Zang[2], W Liu[2] and S Tripathy[2]

[1] Department of Physics, National University of Singapore, 2 Science Drive 3, 117542, Singapore
[2] Institute of Materials Research and Engineering, 3 Research Link, 117602, Singapore

E-mail: zhouhailong@nus.edu.sg and elecsj@nus.edu.sg

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Abstract

Using transmission electron microscopy (TEM), the authors have investigated the behavior of threading dislocations in ZnO selectively grown on a facet-controlled epitaxial overgrown GaN template. In this case, the ZnO is grown by a vapor transport method. The TEM study in the overgrown regions shows that all the pure-edge type dislocations in ZnO are parallel toward the mask area and vertical propagation of dislocation to the ZnO surface is minimized. Using such a selective growth technique on a faceted semi-polar GaN surface, a reduction of threading dislocation density in ZnO could be achieved.

1. Introduction

ZnO has attracted considerable attention over the past years owing to its attractive properties, such as good piezoelectric characteristics, chemical stability, biocompatibility and its potential applications in optoelectronic switches, high-efficiency photonic devices, near-UV lasers and complex three-dimensional nanoscale systems [1–4]. Epitaxial ZnO films have been grown on sapphire by several groups [5, 6] despite the high lattice and thermal mismatch. Recently, several groups reported on the fabrication of n-ZnO/p-GaN electroluminescent devices and demonstrated the potential to realize heterogeneous photonic and electronic devices [7–9]. In such epitaxial heterostructures, the ZnO film quality has been significantly improved because of the lower lattice mismatch ($\sim$1.9%). However, because of the high dislocation density (10^9–10^{10} cm^{-2}) in the GaN grown on c-plane sapphire, the as-grown ZnO films on c-GaN are also expected to contain higher defect densities, which are mainly threading dislocations (TDs). A technique which can help to overcome this problem is facet-controlled epitaxial overgrowth (FACEO) [10–12]. It is realized by starting the epitaxial growth on the (0001) plane and tuning the growth conditions to develop other facets, e.g. $(1\bar{1}01)$ and $(11\bar{2}2)$, on

[3] Authors to whom any correspondence should be addressed.

overgrown stripes. In this context, the ZnO films grown on such FACEO GaN templates are likely to show good crystalline properties with a reduced piezoelectric field at the ZnO/GaN heterointerface. Moreover, from the viewpoint of heterogeneous device integration, it is essential to investigate the propagation of TDs in ZnO overgrown on such FACEO GaN. In this paper, using transmission electron microscopy (TEM), we report on the behavior of TDs in ZnO grown on the FACEO GaN templates. The ZnO growth is carried out by a vapor transport method. The other experimental details for the growth process are available elsewhere [13]. The mechanism behind the reduction of dislocation density in such ZnO is also discussed.

2. Experimental details

Prior to the preparation of the FACEO GaN, a GaN film about 2.0 μm thick was first deposited by metal organic chemical vapor deposition (MOCVD) on a c-plane sapphire substrate. A low-temperature GaN buffer layer was used in this case. Trimethyl gallium and ammonia were used as the sources for Ga and N, respectively, with H_2 as the carrier gas. A 100 nm SiO_2 mask was patterned into stripes oriented in the GaN$\langle 1\bar{1}00\rangle$ direction, defining a 5 μm wide opening with a period of 13 μm. Epitaxial overgrown GaN layers with $\{11\bar{2}2\}$ facets were achieved by controlling the growth temperature and the reactor pressure. The growth temperature and reactor pressure were about 1000 °C and 2.6×10^4 Pa (200 Torr), respectively, to obtain FACEO GaN. Then, these FACEO GaN/sapphire templates were loaded into a tube furnace for the growth of ZnO by thermal vaporization of Zn (99.9% purity) powder and condensation of Zn vapor in the presence of oxygen. The alumina boat with Zn powder was placed at the center of a quartz tube and purged by a helium (99.999% purity) flow of 100 standard cubic centimeters per minute (sccm). The furnace temperature was increased to 800 °C and flow of 99.99% pure oxygen was introduced to the tube reactor at a flow rate of 10 sccm. The mixed O_2 and He gas flows were introduced throughout the whole reaction process, which normally takes 30 min. More detailed growth parameters have been published elsewhere [13]. The cross-sectional morphology of the ZnO/GaN/sapphire layers was studied by using a JSM-6700 field emission scanning electron microscope (SEM) and a Philips CM300 FEG transmission electron microscope (TEM).

3. Structural characterization and discussions

Figure 1 shows cross-sectional SEM images of the FACEO GaN template and the overgrown ZnO grown on top of such GaN templates. The overgrown GaN along the $\langle 1\bar{1}00\rangle_{GaN}$ direction and $\{11\bar{2}2\}$ sidewall facets is clearly displayed in figure 1(a). It is well known that the morphology of such faceted GaN varies with the different growth conditions. In addition, two sets of ZnO/GaN interfacial structures on both sides are observed on the overgrown GaN templates. Besides the original triangular GaN stripes, an extra layer can be seen on top with a uniform thickness. It can be seen that the original faceted GaN stripe has a height of 5 μm and a width of 7 μm. After ZnO overgrowth, near rectangular stripes are observed with a width of about 6.2 μm, which indicates significant lateral growth of ZnO on the FACEO GaN. No extra layers are observed on the SiO_2 mask layer, which further confirms that the ZnO top layer was selectively grown on faceted GaN stripes.

The microstructure of the top layer and the nature of growth of ZnO on FACEO GaN are further investigated by high-resolution TEM (HRTEM). Figure 2 shows a typical HRTEM image of such a ZnO/GaN interface, from which it can be seen that the lattice fringes of ZnO are perfectly aligned with those of GaN and the interface is quite sharp. The corresponding selective area electron diffraction (SAED) pattern is shown in the inset of figure 2. Only one

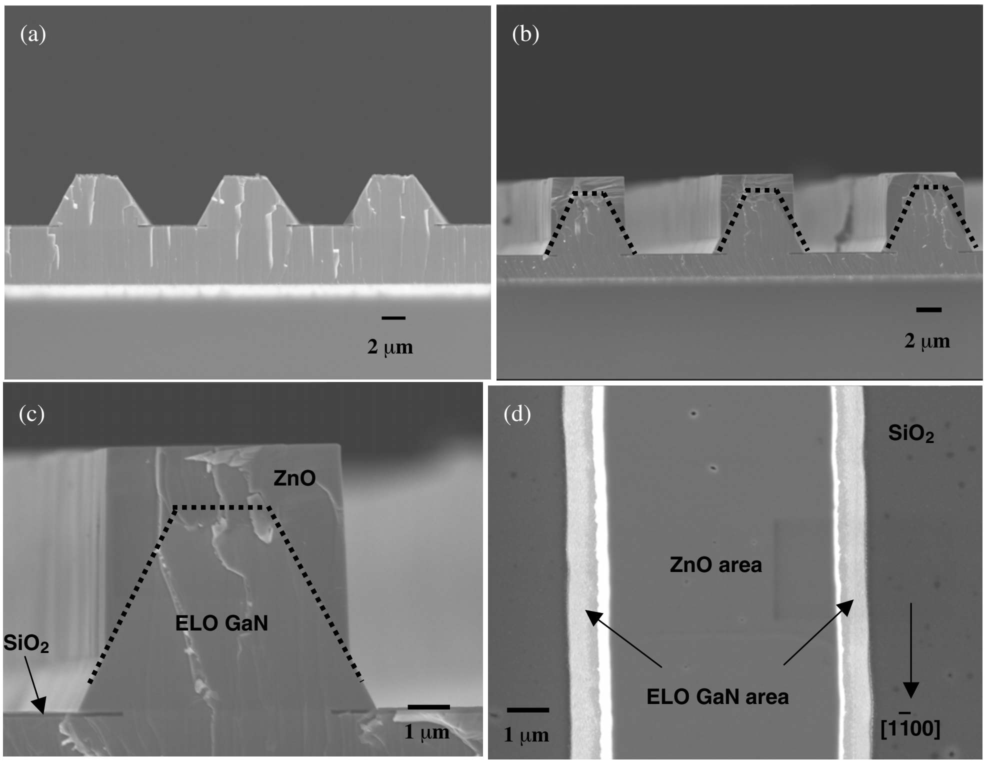

Figure 1. (a) Cross-sectional SEM image of an epitaxial overgrown FACEO GaN template, the apparent asymmetric shapes of the features were induced by the non-perfect perpendicular observation angle. (b) Cross-sectional SEM image of the ZnO/FACEO GaN on sapphire substrate. (c), (d) Cross-sectional and top view SEM images with larger resolution. The ZnO/GaN interface is indicated by the dashed line.

set of SAED pattern is observed, which is due to the very close lattice parameters between wurtzite ZnO and GaN structures. The pattern also verifies the perfect epitaxial growth of high-quality ZnO on MOCVD grown GaN. The cross-sectional TEM images of such ZnO/GaN heterointerfaces with a lower magnification are shown in figure 3. The nature of the dislocations of epitaxial overgrown GaN has been studied by several groups [14–16] and the formation of the horizontal dislocations (HDs) is very important due to the fact that HDs can dramatically decrease the density of TDs in the regrown GaN regions. In general, TDs with Burgers vectors $\mathbf{b} = [0001]$ (screw c-type dislocation), $\mathbf{b} = 1/3[11\bar{2}0]$ (edge a-type dislocation), and $\mathbf{b} = 1/3[11\bar{2}3]$ (mixed $a + c$-type dislocation) were observed in wurtzite epitaxial GaN layers [17]. Since such ZnO on FACEO GaN shows the same wurtzite crystal structure, we have also determined the nature of dislocations in ZnO by a classical two-beam analysis similar to the case of GaN. If a few partial dislocations are present in the GaN layer, a set of two beam images (0002 and $11\bar{2}0$) are sufficient to estimate the different types of Burgers vectors (figure 3). For the condition with the electron beam direction along 0002, both screw-type and mixed-type dislocations can be seen, whereas for the case of a $11\bar{2}0$ beam direction, only edge-type and mixed-type dislocations are observed, based on the out of contrast extinction criterion $\mathbf{g} \cdot \mathbf{b} = 0$. Figure 3(a) is the bright-field image near the interface of the ZnO/FACEO GaN with g vector $11\bar{2}0$. Figures 3(b) and (c) are the dark-field images at the same position

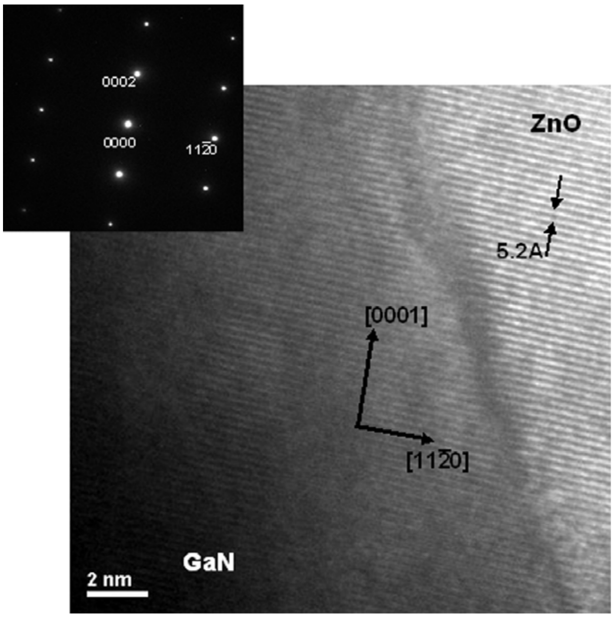

Figure 2. The HRTEM image and the corresponding SAED pattern of the ZnO/FACEO GaN interface.

as the bright-field image shown in figure 3(a) with diffraction spots of [11$\bar{2}$0] and [0002], respectively. The dislocations bent toward the mask areas along the [11$\bar{2}$0] direction are visible in figure 3(c) (type A) but are not visible in figure 3(b). On the other hand, the dislocations with $\mathbf{b}$ = [0001] are also out of contrast in figure 3(b) (type B) but they are clearly visible in figure 3(c). Similar results were also observed by several researchers in MOVPE and hydride vapor phase epitaxy (HVPE)-grown GaN [18, 19]. From this case of invisibility, we have found out that in the regrown ZnO layers the mixed dislocations $\boldsymbol{a} + \boldsymbol{c}$ which split into pure edge dislocations were bent toward the [11$\bar{2}$0] direction. In addition, dislocations with Burger vector of $\boldsymbol{a}$ remain parallel to the c-direction and stop at the FACEO GaN top surface (type C), but no such dislocations propagate to the laterally overgrown ZnO (figure 3(d)). Half of the overgrown ZnO region is shown in figure 3(d), which is also a dark-field image at [0002] diffraction spots. We have observed that bending of dislocations also takes place at the laterally overgrown heterointerfaces and no vertical dislocations go through the ZnO overgrown layer toward the surface. At the same time, lots of dislocations can be observed near the ZnO/GaN interface; not all of them are propagated parallel from the GaN into the ZnO and most of them are generated due to the lattice mismatch induced misfit strain. Based on the previous work of Holec and Srinivasan [20, 21], our ZnO/GaN structures are similar to the single-layer InGaN/GaN system. Compared to the results of Holec's calculations using the slip system proposed by Srinivasan, the critical thickness h_c of the ZnO grown on the GaN surface is about 10 nm, so our ZnO grown on the ELO GaN structures will generate misfit dislocation at the ZnO/GaN interface to relax the strain. These observations reveal that the overall dislocation density on the top surface of ZnO will decrease and thus, with continuous ZnO regrowth, ZnO of high crystal quality can be achieved by this growth method.

The mechanism of dislocation bending can be understood by considering the energy of dislocation lines emerging from a free surface of a crystal [22]. From the view point of

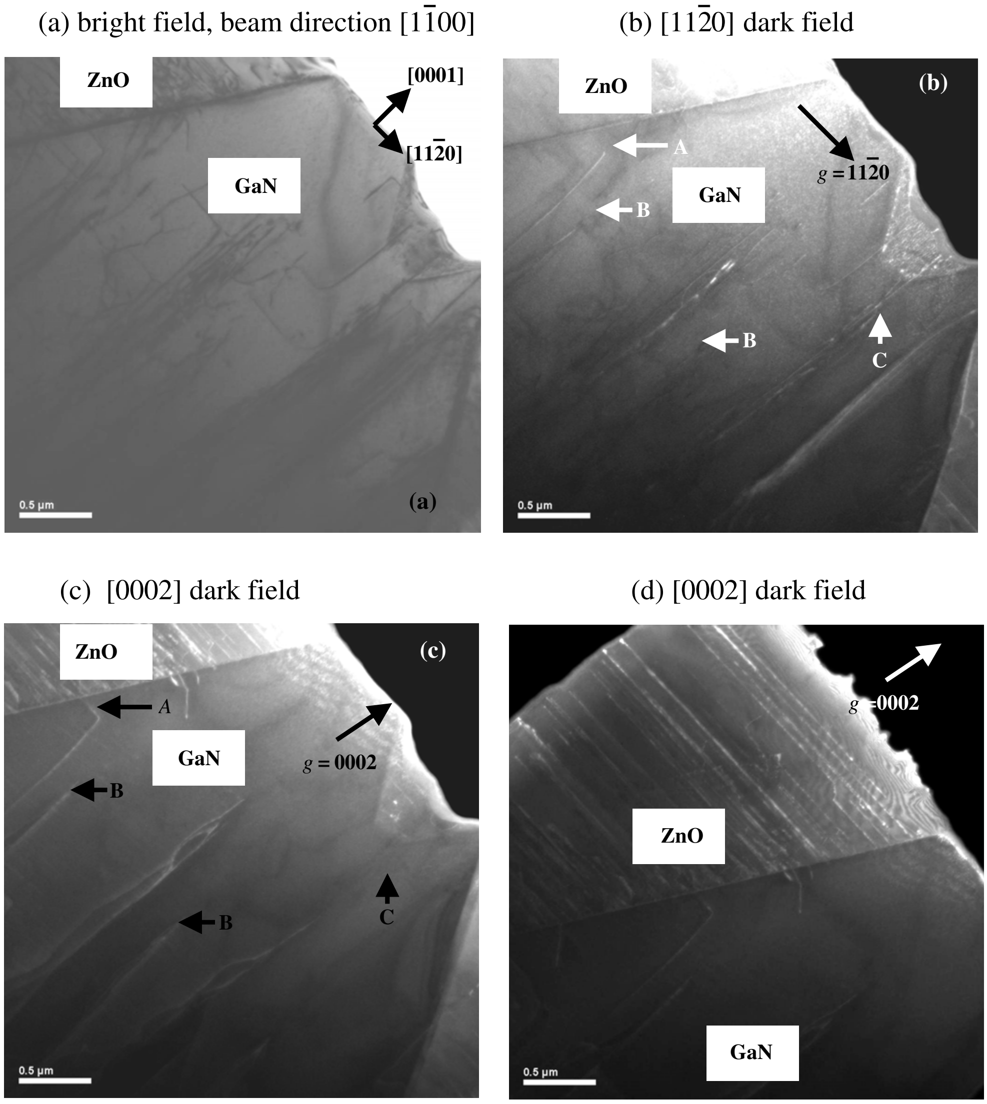

Figure 3. The cross-sectional TEM bright-field image (a) and dark-field images with **g** = 11$\bar{2}$0 (b) and **g** = 0002. (c), (d) Show different locations at the same rectangular ridge ZnO/GaN interface area. TDs show two different bending directions.

dislocation line tension, any dislocation would tend to become perpendicular to a free surface to diminish its energy. As a result, dislocations would gradually change their line directions toward the normal direction of the current facet plane. With different growth modes before and after ZnO overgrowth, capillary stress would induce an interface tension and the imbalance of the interface tension could lead to a dislocation bending mechanism [23] as seen in figure 3. Even for the lattice matched GaN and ZnO structures, such interface characteristics will induce a high density of HDs in ZnO compared to the FACEO GaN template. However, vertical dislocations threading from the ZnO/GaN interface could be minimized. The high quality ZnO wing regions realized by such an overgrowth method suggest that ZnO films can be pseudomorphically grown on the faceted GaN stripes. The HRTEM studies further demonstrate the suitability of overgrown GaN templates for epitaxial regrowth of ZnO and heterogeneous photonic integration.

4. Conclusions

In summary, the mechanism of reduction of dislocation density in ZnO overgrown on FACEO GaN layers has been investigated by HRTEM. Screw-type and mixed-type dislocations in ZnO show a bending toward the mask areas. On the other hand, pure edge and mixed-type dislocations also show a bending in the direction of the mask stripe. The TEM results show that dislocations originating from the ZnO/GaN interface do not propagate to the top surface. Thus, we could achieve a reduction in the overall dislocation density on the ZnO surface. This study shows that epitaxial overgrown GaN templates are suitable for the realization of ZnO with a low dislocation density. In addition, the matching of stacking order, thermal and optical properties and heterostructures with sharp interfaces may provide new opportunities for the fabrication of hybrid ZnO/GaN optoelectronic devices on sapphire substrates.

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References

[1] Xia Y N, Yang P D, Sun Y, Wu Y, Mayers B, Yin Y, Kim F and Yan H 2003 *Adv. Mater.* **15** 353
[2] Pan Z W, Dai Z R and Wang Z L 2001 *Science* **291** 1947
[3] Hu J, Odom T W and Lieber C M 1999 *Acc. Chem. Res.* **32** 435
[4] Chen C S, Kuo C T, Wu T B and Lin I N 1997 *Japan. J. Appl. Phys.* 1 **36** 1169
[5] Kawasaki M, Ohtomo A, Koinuma H, Sakurai Y, Yoshida Y, Tang Z K, Yu P, Wang G K L and Segawa Y 1998 *Mater. Sci. Forum* **264** 1459
[6] Bagnall D M, Chen Y F, Zhu Z, Yao T, Koyama S, Shen M Y and Goto T 1997 *Appl. Phys. Lett.* **70** 2230
Srikant V, Sergo V and Clarke D R 1995 *J. Am. Ceram. Soc.* **78** 1931
[7] Vispute R D, Talyansky V, Choopun S, Sharma R P, Venkatesan T, He M, Tang X, Halpern J B, Spencer M G, Li Y X, Salamanca-Riba L G, Iliadis A A and Jones K A 1998 *Appl. Phys. Lett.* **73** 348
[8] Alivov Y I, Van Nostrand J E, Look D C, Chukichev M V and Ataev D M 2003 *Appl. Phys. Lett.* **83** 2943
[9] Rogers D J, Hosseini Teherani F, Yasan A, Minder K, Kung P and Razeghi M 2006 *Appl. Phys. Lett.* **88** 347
[10] Hiramatsu K, Nishiyama K, Onishi M, Mizutani H, Narukawa M, Motogaito A, Miyake H, Iyechika Y and Maeda T 2000 *J. Cryst. Growth* **221** 316
[11] Miyake H, Narukawa M, Hiramatsu K, Naoi H, Iyechika Y and Maeda T 2001 *Phys. Status Solidi* a **188** 725
[12] Nishizuka K, Funato M, Kawakami Y, Fujita S, Narukawa Y and Mukai T 2004 *Appl. Phys. Lett.* **85** 3122
[13] Zhou H L, Chua S J, Pan H, Zhu Y, Liu W, Zang K Y, Feng Y and Sow C H 2007 *J. Phys. Chem.* C **111** 6405–10
[14] Zheleva T S, Nam O-H, Bremser M D and Davis R F 1997 *Appl. Phys. Lett.* **71** 2472
[15] Kuwano N, Horibuchi K, Kagawa K, Nishimoto S and Sueyoshi M 2002 *J. Cryst. Growth* **237–239** 1047
[16] Ishida M, Ogawa M, Orita K, Imafuji O, Yuri M, Sugino T and Itoh K 2000 *J. Cryst. Growth* **221** 345
[17] Rouviere J L, Arlery M, Daudin B, Feuillet G and Briot O 1997 *Mater. Sci. Eng.* B **50** 61
[18] Sakai A, Sunakawa H and Usui A 1998 *Appl. Phys. Lett.* **73** 481
[19] Honda Y, Iyechika Y, Maeda T, Miyake H and Hiramatsu K 2001 *Japan. J. Appl. Phys.* 2 **40** L309
[20] Holec D, Costa P M F J, Kappers M J and Humphreys C J 2007 *J. Cryst. Growth* **303** 314
[21] Srinivasan S, Geng L, Ponce F A, Narukawa Y and Tanaka S 2003 *Phys. Status Solidi* c **0** 2440
[22] Amelinck S 1982 *Dislocations in Solids* vol 6, ed F R N Nabarro (Amsterdam: Elsevier) pp 67–460
[23] Sakai A, Sunakawa H, Kirmura A and Usui A 2000 *Appl. Phys. Lett.* **76** 442