Journal of Achievements in Materials and Manufacturing Engineering

VOLUME 18
ISSUE 1-2
September-October
2006

Defect structures in InGaN/GaN multiple quantum wells on Si(111) substrates

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Received 15.03.2006; accepted in revised form 30.04.2006

Materials

ABSTRACT

Purpose: Nitrides are compound semiconductor nanomaterials that are suitable for use in light-emitting diodes. It has been desired to grow high quality gallium nitride crystal thin film on silicon substrates because silicon substrates have the advantages of low cost, large wafer size, and good electrical and thermal conductivity. However, the higher defect density can limit the industrial applications due to lower quantum efficiency. The purpose of this study has been to investigate the crystal defect structure within InGaN/GaN multiple quantum wells on Si(111) substrates. In addition, the variation in quantum well thickness was also explained by the selective area growth model.

Design/methodology/approach: InGaN/GaN nano-structures were prepared by metal-organic chemical vapor phase epitaxy (MOVPE) using composite buffer layers. The crystal defect structures in the buried multiple quantum wells on both (0001) and {10-11} sidewalls were carefully studied by transmission electron microscopy. Previous studies on sapphire substrates have been compared and discussed.

Findings: The V defect structures have been found in InGaN/GaN multiple quantum wells on Si(111) substrates. A simplified structural model with increasing barrier thickness has been reported. The barrier thickness increased on both (0001) and {10-11} facets along thin film growth. A decreased fill factor based on the selective area growth model was proposed. In addition, the average thin film growth rate was found to be four times higher along (0001) than that along {10-11} facet. As the number of multiple quantum wells increased, the barrier thickness increasing was also intensified.

Research limitations/implications: The understanding in defect structure could help to modify the processing and design parameters.

Originality/value: The V-defect structure and model were reported for the first time using silicon substrates. The different growth rates along defect structures were quantified. High quality gallium nitride crystal could be manufactured along with better substrate design.

Keywords: Nanomaterials; Electron microscopy; Defect structure; Gallium nitrides

1. Introduction

Gallium nitride-based III–V semiconductors have become promising materials for short-wavelength opto-electronic devices because of their large and direct energy band gaps. The nitride compounds deposited on silicon (Si) substrates have great advantages including lower cost, excellent wafer quality, large wafer size, and good electrical and thermal conductivity. They are also much easier to be integrated with the well-established Si processing technologies [1,2].

However, the production of GaN crystal thin film on silicon substrates still encounters some challenges. It is difficult to grow high quality GaN crystal thin film on Si substrates for commercial applications due to the large difference in lattice constant (17%), crystal structure, and thermal expansion coefficient (57%) that can result in high dislocation density [3-5].

Recently, V defects have received much attention on their structures and formation mechanisms on sapphire substrates. Different structural models have been proposed. The first model, proposed by X. H. Wu et al [6], showed that V defect sidewalls $\{10\bar{1}1\}$ included quantum wells (QWs) and that the space within V defects was empty. Later, N. Duxbury et al [7] reported a similar result based on a spatially resolved energy dispersive x-ray analysis. K. Watanabe et al [8] used HAADF (high angle annual dark field) STEM to study sidewalls including quantum wells and built a similar model. The second model was proposed by N. Sharma et al [9], suggesting V defect sidewalls without InGaN using energy-filtered TEM (EFTEM), which was contradistinctive to the first model. Literatures mentioned above were all based on sapphire substrates. Thus far, no investigation of V defect structures on Si substrate has been reported.

In this study, light-emitting diode (LED) structures with different numbers of QWs (3, 5, and 10) were grown on Si(111) substrates to investigate the defect structures by scanning electron microscopy (SEM) and transmission electron microscopy (TEM). We examined the pit density on the GaN surface from the defects and the barrier thickness variation of multiple quantum wells (MQWs) on both (0001) and $\{10\bar{1}1\}$ facets. The growth rates of MQW epi-layer were also carefully quantified. The correlation between the V defect structure and the barrier thickness was discussed.

2. Experimental

2.1. Materials and LED structure

Epilayers were grown on Si(111) substrates by metal-organic chemical vapor phase epitaxy (MOVPE) using an Aixtron AIX-200/RF-S reactor system. Si(111) was chosen due to its trigonal symmetry favoring epitaxial growth of the GaN(0001) plane. The substrates were cleaned by 5% HF solution prior to the epitaxial growth. Trimethylaluminum (TMAl), trimethylgallium (TMGa), trimethylindium (TMIn) and ammonia (NH_3) were used as Al, Ga, In and N sources, respectively. Monosilane (SiH_4) and hydrogen (H_2) were used as n-type dopant and carrier gas. The samples consisted of GaN sandwiched by two composite buffer layers (CBL) [10] (i.e., Si/AlN/AlGaN/GaN/AlN/AlGaN), n-type GaN:Si, MQWs (InGaN/GaN), and finally p-type GaN:Mg capping layer. The total epitaxy film thickness was about 0.78 µm. The sample has been free of surface crack. The schematic illustration of the sample structure is shown in Fig. 1.

2.2. TEM analysis

The AlN/AlGaN low temperature (LT) buffer layer was grown at 720℃ and the GaN epilayer was grown at 1050℃. The reactor pressure was about 100 mbar. We grew three sets of samples with different numbers in QWs, 3, 5, and 10 respectively. All the process conditions were kept nominally the same. The TEM images of the samples were taken using Philips Tecnai F-20 TEM at 200 kV. Tripod-like polishing tools, followed by argon ion milling to make a thin-enough area, prepared the cross-sectional TEM samples.

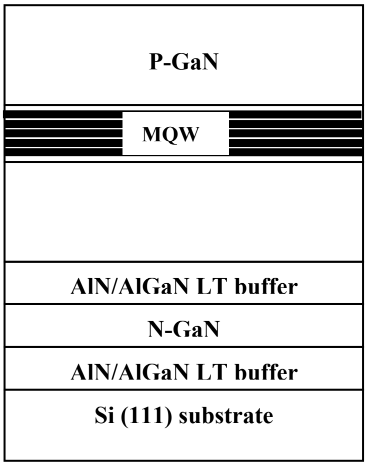

Fig. 1. The schematic illustration of the LED sample structure

3. Results and discussion

3.1. Defect structure

In Fig. 2, the 10-pair MQWs cross-sectional TEM micrograph shows that the V defects include several obvious stripes parallel to the sidewalls $\{10\bar{1}1\}$. Two threading dislocations (TDs) clearly connect to the V defects, with the third (in the right) nearly invisible possibly by the g.b=0 criterion. The stripes parallel to the sidewalls $\{10\bar{1}1\}$ of the V defects correspond with the models of X. H. Wu and K. Watanabe. In Fig. 3, there are no apparent accompanied TDs stopping at the apex of V defects. We found V defects originating from InGaN wells, possibly due to the expansion strain caused by In-rich segregation, and a nucleation site was easily formed. From TEM estimation, the angle between the two $\{10\bar{1}1\}$ facets is about 54.6°±0.5°, which is in close agreement with the theoretical value of 56.1°. Fig. 4 shows the ideal structure model from Fig. 2.

3.2. Barrier thickness

We measured the barrier thickness from the 6th to 9th barriers in Fig. 2 and plotted the data in Fig. 5. It showed that the barrier thickness increased gradually, not only on (0001) but also on the sidewall $\{10\bar{1}1\}$ facets, and the thickness ratio between (0001) and $\{10\bar{1}1\}$ tended to increase intensely. These thickness variations had not been expected, because the process conditions of every barrier layer had been the same. It was likely due to such factors as strain, growth rate, or V defects.

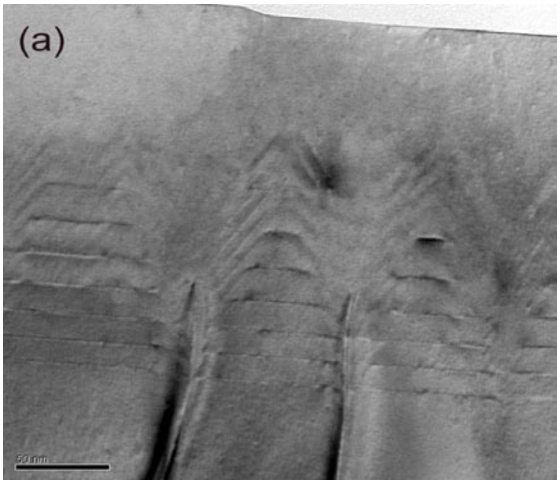

Fig. 2. TEM images of two threading dislocations (TDs) connecting to V defects

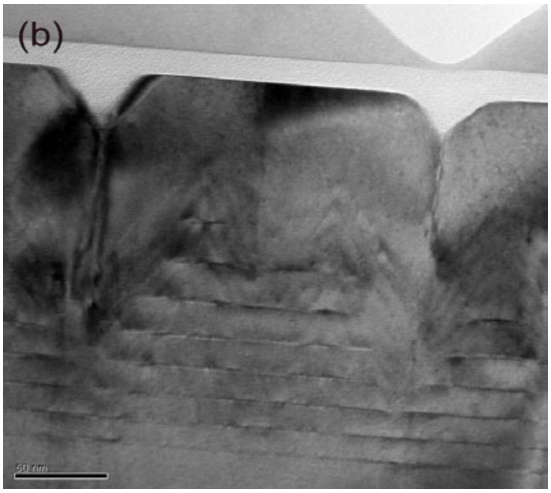

Fig. 3. TEM images of V defects originated from InGaN wells

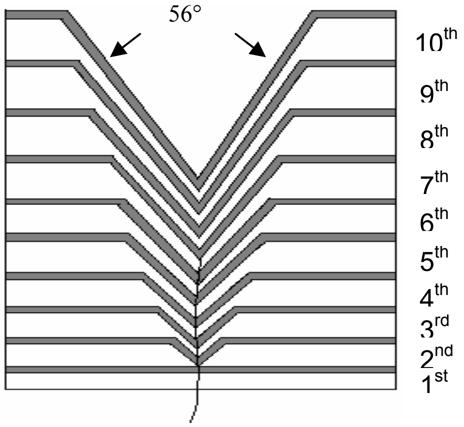

Fig. 4. V defect structure model with increasing barrier thickness

On sapphire substrate, some literatures had investigated well or barrier thickness variations. Sun et al [11] and Im et al [12] have addressed the well thickness increase due to strain relaxation when the QWs number was 10 and accompanied by V defects.

On the other hand, S. Mahanty et al [13] achieved a different result where the quantum well thickness decreased during MQWs growth (from bottom to top), and this was also accompanied by V defects. It was explained that this thickness decreasing phenomenon was due to the cumulative strain caused by lattice mismatch between InGaN and GaN. This slowed down the growth rate and probably reduced In incorporation in the sidewall in comparison with the (0001) plane. To the present, there is no definite conclusion concerning the variations in MQWs thickness and the formation mechanisms. Nevertheless, all these structures had V defects within MQWs.

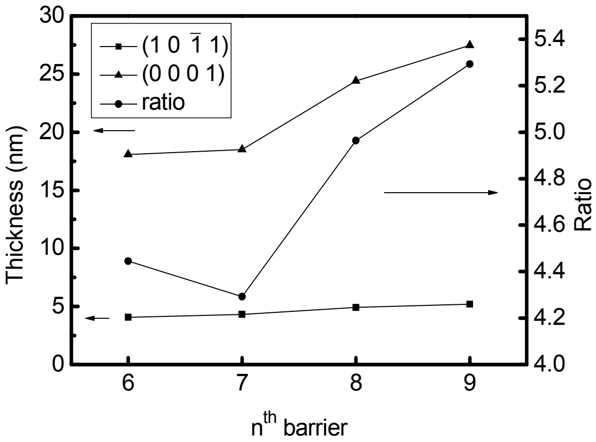

Fig.5. The barrier thickness variation along (0001) and $\{10\bar{1}1\}$ facets and their ratios

3.3. Growth rate

From the TEM image, the average growth rate on (0001) and $\{10\bar{1}1\}$ facets was estimated to be about 1 and 0.24 nm/min, respectively. The data suggested that the growth rate depended on crystalline planes. It showed a slower growth rate in the V defect region both in the well and in the barrier. It has been reported that In or Ga incorporation (implying growth rate) on $\{10\bar{1}1\}$ facets was slower than that on (0001). The empty area of V defects became larger gradually when MQWs grew from the bottom to the top, while the residual area (i.e., the total area at well or barrier layer minus the area inside V defects) available for precursors to deposit became less. Additionally, the growth rate of MQW epi-layers gradually increased from the bottom to top layer. We supposed that there was little destruction of reactants in V defect regions, and therefore an extra supply of precursors was available for growth in the near-by regions. This result was very similar to that in SAG (selective area growth) phenomenon. Therefore, it resulted in an enhancement of growth rate outside the V defect regions when the fill factor was decreased.

3.4. Effects of quantum well number

Fig. 6 shows the remarkable tendency for the barrier thickness to increase in the 3, 5 and 10 pairs of QWs, all along (0001). In 3-QWs, the 3rd barrier was 10% thicker than the 2nd barrier. In 5-QWs, the 5th barrier was 28% thicker than the 2nd barrier. In 10-QWs, the 9th barrier thickness was more than double (2.3 times) that of the 2nd barrier. We also measured the well thickness to be about 1.1±0.2 nm, but no apparent variation trend appeared. When the number of QWs increased, more V defects were also formed, causing the barrier thickness to cumulate intensely.

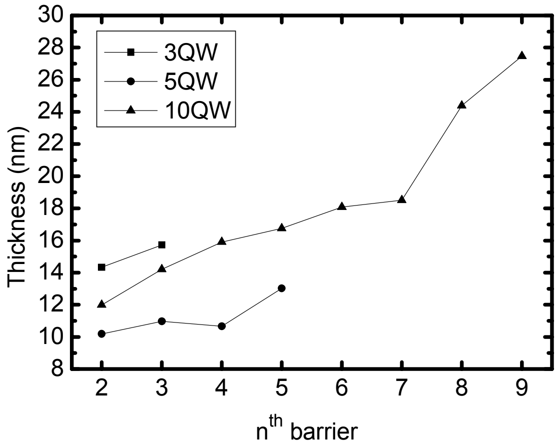

Fig. 6. The barrier thickness in the 3, 5, and 10 pairs of MQW samples

4. Conclusions

In summary, the V defect structure has been found to have the buried InGaN/GaN on sidewall $\{10\bar{1}1\}$ in MQWs on Si(111) substrate. This is more in accordance with the models by X. H. Wu and K. Watanabe. A simplified structure model with increasing barrier thickness has thus been proposed. The barrier thickness of MQWs gradually increased on both (0001) and $\{10\bar{1}1\}$ facets along deposition, which were revealed by TEM. The average thin film deposition growth rate was quantified to be four times higher along (0001) than along $\{10\bar{1}1\}$ facet. As the number of QWs increased, the barrier thickness increase also intensified.

Acknowledgements

This work was partially supported by the National Science Council under research grant NSC92-2745-L182-001.

References

[1] A. Cahill and M. A. El Baradie, LED-based fibre-optic sensor for measurement of surface roughness, Journal of Material Processing Technology, 119 (2001) 299-306.

[2] A. N. Gulluoglu and C. T. Tsai, Dislocation generation in GaAs crystals grown by the vertical gradient freeze method, Journal of Material Processing Technology, 102 (2000) 179-187.

[3] N. Subramanyam and C. T. Tsai, Dislocation reduction in GaAs crystal grown from the Czochralski process, Journal of Material Processing Technology, 55 (1995) 278-287.

[4] G. M. Wu and T. H. Hsieh, Crystal quality and electrical properties of p-type GaN thin film on Si(111) substrate by metal-organic chemical vapor deposition MOCVD, Journal of Material Processing Technology, (2006) in press.

[5] G. Cong, Y. Lu, W. Peng, X. Liu, X. Wang and Z. Wang, Design of the low temperature AlN interlayer for GaN grown on Si (111) substrate, Journal of Crystal Growth, 276 (2005) 381-388.

[6] X. H. Wu, C. R. Elsass, A. Abare, M. Mack, S. Keller, P. M. Petroff, S. P. Denbaars, J. S. Speck and S. J. Rosner, Structural origin of V-defects and correlation with localized excitonic centers in InGaN/GaN multiple quantum wells, Applied Physics Letters, 72 (1998) 692-694.

[7] N. Duxbury, U. Bangert, P. Dawson, E. J. Thrush, W. van der Stricht, K. Jacobs and I. Moerman, Indium segregation in InGaN quantum-well structures, Applied Physics Letters, 76 (2000) 1600-1602.

[8] K. Watanabe, J. R. Yang, S. Y. Huang, K. Inoke, J. T. Hsu, R. C. Tu, T. Yamazaki, N. Nakanishi and M. Shiojiri, Formation and structure of inverted hexagonal pyramid defects in multiple quantum wells InGaN/GaN, Applied Physics Letters, 82 (2003) 718-720.

[9] N. Sharma, P. Thomas, D. Tricker and C. Humphreys, Chemical mapping and formation of V-defects in InGaN multiple quantum wells, Applied Physics Letters, 77 (2000) 1274-1276.

[10] S. J. Lee, S. H. Jang, S. S. Lee and C. R. Lee, Growth and characterization of $In_{0.28}Ga_{0.72}N$/GaN multiple-quantum wells on Si(111), Journal of Crystal Growth, 249 (2003) 65-71.

[11] C. J. Sun, M. Z. Anwar, Q. Chen, J. W. Yang, M. A. Khan, M. S. Shur, A. D. Bykhovski, Z. Liliental-Weber, C. Kisielowski, M. Smith, J. Y. Lin and H. X. Xiang, Quantum shift of band-edge stimulated emission in InGaN-GaN multiple quantum well light-emitting diodes, Applied Physics Letters, 70 (1997) 2978-2980.

[12] J. S. Im, H. Kollmer, J. Off, F. Scholz and A. Hangleiter, Carrier confinement in GaInN/AlGaN/GaN quantum wells with asymmetric barriers: direction of the piezoelectric field, Materials Science and Engineering B: Solid-State Materials for Advanced Technology, 59 (1999) 315-318.

[13] S. Mahanty, M. Hao, T. Sugahara, R. S. Q. Fareed, Y. Morishima, Y. Naoi, T. Wang and S. Sakai, V-shaped defects in InGaN/GaN multiquantum wells, Materials Letters, 41 (1999) 67-71.