Extended Abstracts of the 2009 International Conference on Solid State Devices and Materials, Sendai, 2009, pp893-894

Growth of Quaternary AlInGaN with Various TMI Molar Rates

Sheng-Fu Yu, Shoou-Jinn Chang and Sheng-Po Chang

Institute of Microelectronics & Department of Electrical Engineering
Center for Micro/Nano Science and Technology
Advanced Optoelectronic Technology Center
National Cheng Kung University, Tainan 70101, Taiwan
Phone: +886-6-2757575-62400-1208 E-mail: adolph0207@gmail.com

1. Introduction

Ultraviolet LEDs use GaN as the material for well layers and use AlGaN as the materials for barrier layers. Thus, similar piezoelectric field-induced QCSE will reduce the intensity of the LED due to the lattice mismatch between GaN and AlGaN. Recently, it has been shown that the quaternary AlInGaN permits an extra degree of freedom by allowing independent control of the band gap and lattice constant. The value of this freedom is the ability to engineer lattice-matched heterostructures or novel structures with different layers subjected to adjustable degree of in-plane stress. By replacing InGaN and AlGaN with lattice matched AlInGaN, we should be able to totally eliminate piezoelectric field-induced QCSE in GaN-based LEDs. However, the optical and electrical properties of the quaternary AlInGaN are still relatively unknown [1, 2]. This is due to the complicated mechanism as indium and aluminum incorporated into the GaN epilayer simultaneously. It is known that high quality AlGaN should be grown at high temperature in H_2 environment while high quality InGaN should be grown at low temperature in N_2 environment. The large differences in growth temperature and lattice constant of the binary compounds might also result in severe phase separation. As a result, it is difficult to achieve high quality AlInGaN epilayers with enough thickness (~100 nm), which is important for the understanding of its fundamental properties [3, 4]. In this paper, we report the growth of very thick AlInGaN epilayers (~400 nm) on GaN/sapphire template. Detailed growth parameters and the properties of the thick AlInGaN epilayers will also be discussed.

2. Experiments

Samples used in this study were all grown on c-plane sapphire substrate by atmospheric pressure metalorganic chemical vapor deposition (MOCVD). During the growth, trimethylaluminum (TMA), trimethylgallium (TMG), trimethylindium (TMI), and ammonia (NH_3) were used as the precursors of Al, Ga, In and N, respectively. A 25-nm-thick low-temperature GaN nucleation layer at 550°C and a 3-μm-thick undoped GaN buffer layer at 1130°C using H_2 as the carrier gas. The quaternary AlInGaN layer was subsequently grown on the GaN/sapphire template at 850°C for two hours using N_2 as the carrier gas. During the growth of AlInGaN, we kept the flow rates of TMA, TMG and NH3 at 2.6 μmol/min, 7.0 μmol/min and 0.22 mol/min. On the other hand, the TMI molar flow rate was kept at **0**, **9.38**, **18.76** and **37.53** μmol/min for samples 1, 2, 3 and 4, respectively.

After the growth, a PANalytical X'Pert pro MRD high resolution x-ray diffraction (HRXRD) Reciprocal space mapping (RSM) measurements were also performed to determine lattice constant of the AlInGaN epilayers and the strain effect. The electronic micro probe analysis (EMPA) was also used to determine the amount of aluminum and indium incorporations. Surface morphologies of the samples were then evaluated by a field-emission scanning electron microscopy (FESEM). Hall measurements were also performed to study the electrical properties of the as-grown samples.

3. Results and discussion

To investigate effects of the incorporation of Al and In, XRD RSM measurements were performed on the asymmetric (1 0 5) crystal planes. As shown in figure 1, it was found that the main GaN peaks and the AlInGaN peaks were located at the same axis q_y. This indicates that these four samples were all full strained without any relaxation along the in-plane direction. On the other hand, it was found that the out-of-plane strain changed from tensile to compressive as we increased the TMI molar flow rate from 0 to 37.53 μmol/min. It should be noted that lattice constant of the AlInGaN layer prepared with TMI molar flow rate of 18.76 μmol/min (i.e., sample 3) matched perfectly with the underneath GaN buffer layer.

Table I Summarized (302) FWHM, EPMA measurement, mobility and concentration results with various TMI molar flow rate

Sample	Mobility (cm2/V-s)	Concentration (1/cm3)	EPMA Al	EPMA In	(302) FWHM (aresec)
			composition (%)		
1	802	3.47E17	13	------	504
2	457	4.55E17	11.1	1.02	496.8
3	236	5.063E17	8.86	2.01	453.6
4	120	6.753E17	7.586	3.01	626.4

Table I summaries Al and In composition ratios obtained from EPMA measurement, and the results of Hall measurements for the four samples. It was found that In content in the AlInGaN epilayers increased as we increased the TMI molar flow rate. However, Al content decreased although the TMA molar flow rate was kept constant. Similar results have also been reported by Liu et al [5]. It was also found that the In content doubled from 1.02% to 2.01% as we doubled the TMI molar flow rate from 9.38 to 18.76 μmol/min. However, it only increased to 3.01% as we doubled the TMI molar flow rate again to 37.52 μmol/min. This should be attributed to the saturation of In incorporation rate at 850°C in N_2 ambient. It was also found that the electron concentration in the AlInGaN epilayers increased while the mobility decreased as we increased the TMI molar flow ratio.

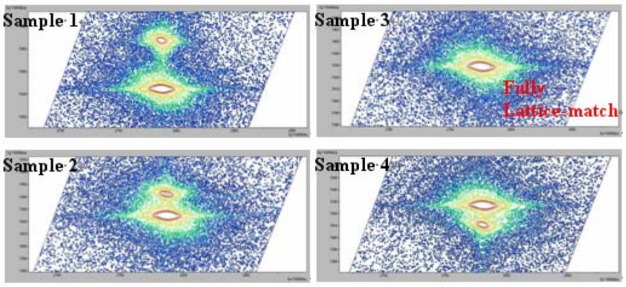

Fig. 1 RSM mapping on the asymmetric (1 0 5) crystal planes for the AlInGaN epilayers prepared in this study.

Figure 2 shows FESEM images of the samples prepared in this study. It was found that there exist numerous inverted hexagonal pits on the surfaces of these samples. From these FESEM images, it was found that averaged pit density were 6.6×10^8, 4.6×10^8, 1.9×10^8, 2.1×10^8 cm^{-2} for samples 1, 2, 3 and 4, respectively. It was also found that pit size depends strongly on the TMI molar flow rate during the growth of AlInGaN. As we increased the TMI molar flow rate from 0 to 18.76 μmol/min, it was found that pit density decreased significantly. The size of the pits also became smaller. It was also found that pit density increased slightly as we further increased the TMI molar flow rate to 37.52 μmol/min. It has been shown previously that these V-shape defect pits were associated with threading dislocations (TDs) in the epilayers [6, 7]. These observation indicates that the number of TD was minimized in sample 3. To further investigate crystal quality of these quaternary films, we performed XRD (002) ω-scans and also measured their XRD (302) rocking curves. It was found that full-width-half-maxima (FWHMs) of the XRD (002) ω-2θ scanned AlInGaN peaks were all around 240 aresec, which suggest good crystal qualities of our films. The measured FWHMs of the XRD (302) rocking curves were listed in Table I. It has been shown previously that (302) ω-scan reflection is an effective way to evaluate crystal quality of GaN-based epilayers [8, 9]. As we increased the TMI molar flow rate from 0 to 18.76 μmol/min, it was found that XRD (3 0 2) FWHM decreased monotonically, and increased as we further increased the TMI molar flow rate to 37.52 μmol/min. These observations agree well with those observed from the FESEM images shown in figure 2. The lowest pit density and the smallest (3 0 2) FWHM observed from sample 3 should be both attributed to the lattice matched AlInGaN which minimized the number of TD.

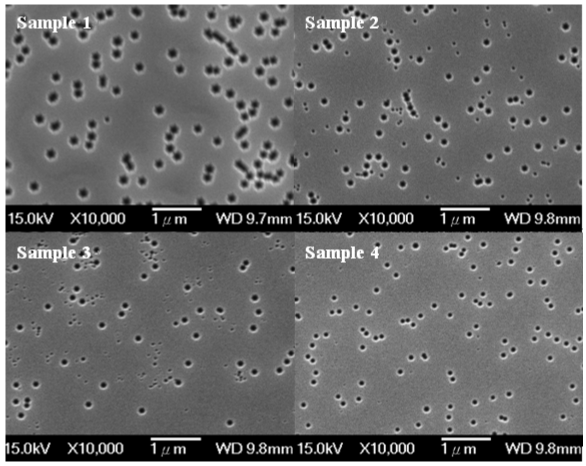

Fig. 2 FESEM images of the samples prepared in this study.

4. Conclusions

In summary, we demonstrated the growth of very thick (~400 nm) quaternary AlInGaN layer on GaN/sapphire template by MOCVD. By properly controlling the TMI molar flow rate, we successfully achieved an $Al_{0.89}In_{0.02}GaN$ layer lattice matched to the underneath GaN buffer. It was found that we can minimize the number of V-defect pits and the XRD (3 0 2) FWHM.

Acknowledgements

This work was supported by National Science Council under contract number NSC 95-2221-E-006-314 and NSC 95-2221-E-006-357-MY3. This work was also supported in part by the Center for Frontier Materials and Micro/Nano Science and Technology, National Cheng Kung University, Taiwan (D97-2700).

References

[1] Y. Liu, T. Egawa, H. Ishikawa, and T. Jimbo, J. Crystal Growth 259 (2003) 245.
[2] Y. Kobayashi, Y. Yamaguchi, and N. Kobayashi, Jpn. J. Appl. Phys. 42 (2003) 2300.
[3] Y. Liu, T. Egawa, H. Ishikawa, B. Zhang, and M. Hao, Jpn. J. Appl. Phys. 43 (2004) 2414.
[4] D. Amabile, R. W. Martin, T. Wang, M.A. Whitehead, and P. J. Parbrook, Phys. Stat. Sol. (c) 7 (2003) 2478.
[5] Y. Liu, T. Egawa, H. Ishikawa, and T. Jimbo, Phys. Stat. Sol. (a) 200 (2003) 36
[6] H. Kang, S. Kandoor, S. Gupta, I. Ferguson, S.P. Guo, and M. Pophristic, Phys. Stat. Sol. (c) 2 (2005) 2145.
[7] H. M. Wang, J. P. Zhang, C. Q. Chen, Q. Fareed, J. W. Yang, and M. Asif Khan, Appl. Phys. Lett. 81 (2002) 4.
[8] R. Chierchia, T. BWttcher, H. Heinke, S. Einfeldt, S. Figge, and D. Hommel, J. Appl. Phys. 93 (2003) 11.
[9] H. Heinke, V. Kirchner, S. Einfeldt, and D. Hommel, Appl. Phys. Lett. 77 (2000) 14.