Extended Abstracts of the 2011 International Conference on Solid State Devices and Materials, Nagoya, 2011, pp1393-1394

Tilted domain and indium content of MOVPE-grown InGaN layer on *m*-plane GaN substrate

Kanako Shojiki[1], Takashi Hanada[1,2], Takaaki Shimada[1], Yuhuai Liu[1,2], Ryuji Katayama[1,2] and Takashi Matsuoka[1,2]

[1] Institute for Materials Research, Tohoku University
2-1-1 Katahira, Aoba-ku, Sendai, Miyagi 980-8577, Japan
Phone: +81-22-215-2281 E-mail: k.shojiki@imr.tohoku.ac.jp
[2] CREST, Japan Science and Technology Agency

1. Introduction

InGaN grown along nonpolar orientations have attracted a great deal of attention, since the strained quantum wells (QWs) fabricated on nonpolar planes are free from the drawback of quantum-confined Stark effects caused by the polarization which reduces the oscillator strength of electron-hole pairs in *c*-plane QWs. However, the indium incorporation into the *m*-InGaN film is much more difficult than that into the *c*-InGaN film grown by metalorganic vapor phase epitaxy (MOVPE). Recently, the effects of miscut angles and their directions [1-3], and resultant low defect densities [4] have been studied to improve the indium incorporation efficiency, crystal qualities and optical properties. In particular, the enhancement of indium incorporation efficiency with increasing the miscut angle of the *m*-GaN substrate has been reported [1,3]. Also in the recent report, a crystallographic tilting which is characteristic of *m*-plane InGaN has been detected [5]. However, unfortunately the indium contents described in the above reports may be inaccurate since there is not enough description on the evaluation process. So in this study, we explored the way to estimate the accurate alloy composition of nonpolar InGaN by X-ray diffraction (XRD) reciprocal space mapping (RSM) measurements, and investigated the effect of miscut angle and growth rate.

2. Experimental

The *m*-plane GaN substrates with different miscut angles toward *c* direction were prepared by slicing the thick *c*-plane GaN crystals which were grown by hydride vapor phase epitaxy (HVPE) at Mistubishi Chemical corporation. InGaN layers were grown by horizontal-type quartz-reactor MOVPE. Triethylgalllium (TEGa), trimethylindium (TM In) and ammonia (NH_3) were used as precursors for gallium, indium and nitrogen, respectively. After the degreasing in the organic solvents and the removal of the surface oxide by dipping into piranha solution for 60 seconds, substrates were heated up for 5 minutes at 900^{o}C under nitrogen ambient as a thermal cleaning, followed by the growth of InGaN layers at 650^{o}C for 120 minutes. The growth temperature was measured by a thermocouple placed just beneath the center of a susceptor. Two groups (A and B) of InGaN samples were grown under different conditions as summarized in Table I.

Table I MOVPE growth conditions of InGaN layers

MOVPE growth condition	group A	group B
reactor pressure (Torr)	650	
V/III ratio	8000	8400
NH_3 flow rate (slm)	15	
TMIn/(TMIn+TEGa)	0.97	0.90
TMI flow rate (μmol/min)	81.3	72.2
TMGa flow rate (μmol/min)	2.8	8.0
miscut angle (degree)	0.0, 2.2, 5.0	0.1, 2.3, 4.7

The film thicknesses were estimated by the curve fitting to the $2\theta/\omega$ profiles of a symmetric-plane diffraction which exhibited clear Pendellösung fringes, aside from the stylus profiler. All the XRD measurements were performed using high-resolution X-ray diffractometer (Bruker-AXS, D8 Discover) equipped with an one-dimensional detector array (VANTEC-1). The indium contents of the InGaN films were determined from the application of the Vegard's law to the relaxed lattice constants. Here the relaxed lattice constants were obtained based on the iteration method using measured strained lattice constants and stiffness parameters in the literature [6]. In this work, two independent RSM measurements were employed in order to obtain three different lattice constants of *a*, *m* and *c* unambiguously, since the *m*-plane films were under triaxial strains. As for the Miller indices of diffraction of interest, $(20\bar{2}1)$ and $(21\bar{3}0)$ were adopted. In these configurations, three lattice constants as well as rotations of the crystals around *a*- and *c*-axis should be detected. In principle, the superposition of the crystallographic rotation makes it hard to determine the lattice strain because the axial shifts of the peak in the reciprocal space may be caused by both effects of the crystallographic rotation and the lattice distortion. However, thanks to the choice of the above special pair of diffractions, the effect of the rotation can be excluded from the strain evaluation process by a simple geometrical correction, since the above two diffractions involve the *m*-directional components with a same length of reciprocal lattice vector. In other words, a certain lattice spacing should be constant even if it is to be observed from other directions. In this study, as a result of the above processes, the indium contents were accurately characterized and we found the novel tilted-domain structures with a characteristic asymmetry.

3. Result and discussion

The film thicknesses of group A were around 150 nm, while those of group B were about twice as group A. Fig. 1 (a,b) show typical RSMs of $(20\bar{2}1)$ and $(21\bar{3}0)$ diffractions from group-B InGaN film on 0.1^o-off GaN substrate. Fig. 1(c,d) shows the XRD rocking curves of InGaN $(20\bar{2}1)$ and $(21\bar{3}0)$ diffraction for group-B samples, which correspond to the lines of intersection between a RSM contours and equi-length surfaces of the reciprocal lattice vector, as depicted by dotted lines in Fig. 1(a) and (b), respectively. It is found that all the $(21\bar{3}0)$ curves of group-B samples split into three peaks as shown in Fig. 2(d). It means that there appeared three domains with different orientations which rotated around *c*-axis toward $\pm a$ directions, which is detectable in case of $(21\bar{3}0)$ diffractions. On the other hand, as for the $(20\bar{2}1)$ diffraction, there was no trace of crystallographic rotation around *a*-axis since there was no additional peak other than a single InGaN peak as shown in Fig. 2(c). It is surprising that the detected tilting direction ($\pm a$) is toward the vertical direction with respect to the miscut direction (*c*), as opposed to the usual case of well known III-V semiconductors on misoriented substrates, where the tilting direction is always parallel to the miscut direction.

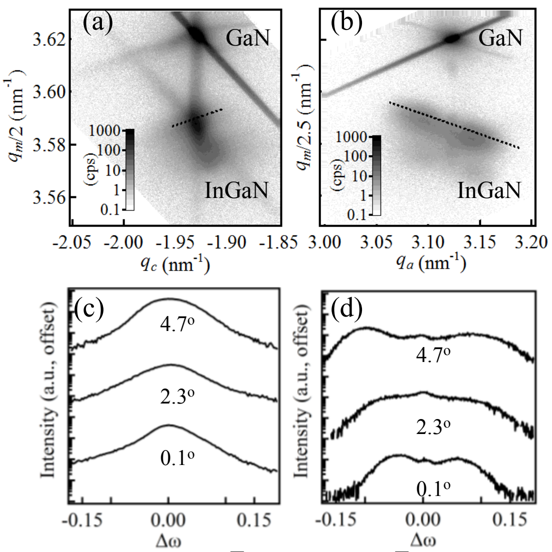

Fig. 1 RSMs of (a) $(20\bar{2}1)$ and (b) $(21\bar{3}0)$ diffractions from group-B InGaN film on 0.1^o-off GaN substrate. Rocking curves for InGaN (c) $(20\bar{2}1)$ and (d) $(21\bar{3}0)$ diffraction.

Moreover, it should be noted that the tilted domains were only observed in group-B samples with higher growth rate which are thicker than group A. This fact implies that the above characteristic tilting behavior is closely related to the lattice relaxation process which happens in case of the films with a thickness beyond the critical thickness. It is already pointed out by our previous study that the introduction of a set of misfit dislocations which gives a tilting toward $\pm a$ direction is easier than toward *c* direction [3].

Fig. 2 shows the miscut angle dependence of the indium content derived from the RSMs data taking into account the tilt and the residual triaxial strain. The increase of the substrate miscut angle leads to the apparent increase of the indium contents. It is provable that the increase of the miscut angle enhance the indium incorporation since it increases the densities of steps and kinks appeared on the growth front, at which the desorption of the indium atoms will be suppressed. Furthermore, group B with higher growth rate revealed the higher indium content than group A. This is explained by the following two steps. Firstly as the growth rate increases, the growth condition reaches to the highly non-equilibrium one, the indium incorporation efficiency increases as in the case of *c*-plane InGaN [7]. Next as the growth proceeds and once the thickness exceeds the critical thickness, the indium incorporation into GaN matrix dramatically enhanced since the compressive strain has been rapidly released, which is often found in III-V materials growths, known as composition pulling effects.

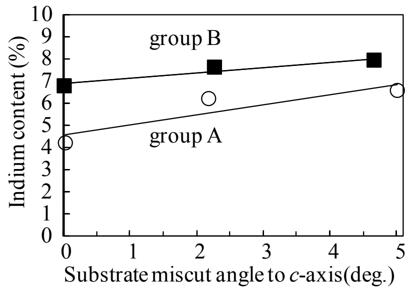

Fig. 2 Miscut angle dependence of indium content.

4. Conclusion

The accurate way to estimate the alloy composition in strained non-polar films with wurtzite crystal structure has been developed based on XRD RSM measurements. The enhancements of the indium incorporation efficiency in InGaN with increasing the miscut angle and growth rate, as well as a novel lattice relaxation process with a three-fold-tilting structure, were found for the first time.

References

[1] H. Yamada, K. Iso, H. Masui, M. Saito, K. Fujito, S. P. Denbaars, and S. Nakamura, J. Cryst.Growth **310**, 4698 (2008).

[2] Y. D. Lin, M. T. Hardy, P. S. Hsu, K. M. Kelchner, C. Y. Huang, D.A. Haeger, R. M. Farrell, K. Fujit, A. Chakraborty, H. Ohta, J. S. Speck, S. P. Denbaars, and S. Nakamura, Appl. Phys. Express **2**, 082102 (2009).

[3] T. Hanada, T. Shimada, S. Y. Ji, K. Hobo, Y. H. Liu, and T. Matsuoka, phys. stat. sol. (c) **8**, 444 (2011).,

[4] S. F. Chichibu, H. Yamaguchi, L. Zhao, M. Kubota, T. Onuma, and K. Okamoto, Appl. Phys. Lett. **93**, 151908 (2008).

[5] S. Yoshida, T. Yokogawa, Y. Imai, S. Kimura, and O. Sakata, *Abstract of Asia-Pacific Workshop on Widegap Semiconductors (APWS-2011),* We-B2, Mie, Japan (2011).

[6] F.M. Morales, D.Gonzalez, J.G. Lozano, R. Garcia, S. Hauguth-Frank, V. Lebedev, V. Cimalla, and O. Ambacher, Acta Mater. **57**, 5681 (2009).

[7] S. Keller, B.P. Keller, D.Kapolnek, A. C. Abare, H. Masui, L. A. Coldren, U. K. Mishra, and S. P. Denbaars, Appl. Phiys. Lett. **68**, 27 (1996).