# Compositional Pulling Effect in Epitaxial Growth of GaInN by RF-MBE

Tomohiro Yamaguchi<sup>1</sup>, Takuo Sasaki<sup>2</sup>, Masamitu Takahasi<sup>2</sup>, Tsutomu Araki<sup>3</sup>, Takeyoshi Onuma<sup>1</sup>, Tohru Honda<sup>1</sup> and Yasushi Nanishi<sup>3</sup>

> <sup>1</sup> Kogakuin University Department of Applied Physics Nakano-machi 2665-1, Hachioji, Tokyo 192-0015, Japan Phone: +81-42-628-4651 E-mail: t-yamaguchi@cc.kogakuin.ac.jp
> <sup>2</sup> National Institutes for Quantum and Radiological Science and Technology (QST) Synchrotron Radiation Research Center 1-1-1 Koto, Sayo, Hyogo 679-5148, Japan
> <sup>3</sup> Ritsumeikan University Department of Electrical & Electronic Engineering 1-1-1 Noji-higashi, Kusatsu, Shiga 525-8577, Japan

#### Abstract

We report on the compositional pulling effect in the epitaxial growth of GaInN by radio-frequency plasma-assisted molecular beam epitaxy (RF-MBE), observed using *in-situ* X-ray diffraction (XRD) reciprocal space mapping (RSM) measurements. The epitaxial growth kinetics and mechanism are also discussed using the results of the evolutions of In composition as well as growth rate of GaInN in the initial growth stage.

#### 1. Introduction

GaInN alloys have been attracting considerable attention for application to not only visible but also infrared light-emitting devices since the band gap of InN was reported in 2002 to be around 0.7 eV. They can cover potentially the most part of solar energy spectrum. This attracted further interest in these materials as extremely high efficiency solar cells. However, the fabrications of high-quality GaInN-based structures, including a  $Ga_{I-x}In_xN$  thick film and a  $Ga_{I-y}In_yN/Ga_{I-x}In_xN$  (y>x) quantum well on the thick film have still been challenging topics, owning to the heteroepitaxial growth with a large lattice mismatch system of about 11 % between GaN and InN. The deeper understanding of the epitaxial growth kinetics and mechanism has been still required.

In this talk, we report on the compositional pulling effect in the epitaxial growth of GaInN by radio-frequency plasma-assisted molecular beam epitaxy (RF-MBE), observed using *in-situ* X-ray diffraction (XRD) reciprocal space mapping (RSM) measurements.

#### 2. Experiments

The *in-situ* RSM measurements were performed using the MBE directly coupled to an X-ray diffractometer at beamline 11XU of the synchrotron radiation facility SPring-8. A GaInN film was grown on a metalorganic vapor phase epitaxy (MOVPE)-grown (0001)GaN/ $\alpha$ -Al<sub>2</sub>O<sub>3</sub> template. The V/III ratio during growth was set to be less than 1 (under a metal-rich condition). The expected In solid composition of GaInN was about 55 % from the supplied ratio between Ga and nitrogen radicals (N\*) in this study [1]. The growth rate of GaInN was approximately 0.08 ML/sec. The two-dimensional (H [10-10] – L [0001] coordinate in Miller indices) RSM around (10-11) diffraction peak was measured by adjusting the sample orientation and the detector position. A RSM image was taken at every 7 sec., that is, GaInN with a thickness of approximately 0.14 nm was grown in a scan.

#### 3. Results and discussion

Figure 1 shows one of RSMs obtained in this study. The evolution of peak position for (10-11) diffraction peak of GaInN as a function of growth time is also shown in Fig. 1. After the growth of approximately 140 sec., the diffraction peak of GaInN was observed. After the appearance of GaInN peak, both the diffraction positions of H and L were shifted in such a direction away from the GaN peak position. After the growth of 300-500 sec., the diffraction position of H was shifted further away from the GaN peak position, while that of L was conversely shifted toward GaN peak position.

Figure 2 shows the evolution of In composition and relaxation ratio of GaInN, estimated from the result of Fig. 1. From the evolution of relaxation ratio, GaInN was found to be gradually relaxed. Now we focus on the evolution of In composition. Up to approximately 400 sec., the In composition increased as GaInN grew. After that, the composition was almost constant of about 55 %, which was the composition expected from the supplied ratio in this growth condition.

Figure 3 shows the evolution of the integrated intensity of GaInN (10-11) peak during growth. The integrated intensity was constantly increased except for the first ~400 sec. This indicates that the growth rate is almost constant in this term. However, it can be seen that the growth rate of GaInN was slow in the initial growth stage up to ~400 sec.

These can be explained by the fact that not only lattice

relaxation but also compositional pulling effect [2] at the initial growth stage took place as means of reducing the strain of GaInN. The low growth rate of GaInN at the initial growth stage might be due to desorption of nitrogen.

The growth of GaInN on GaN is in the compressive strain state. The result of the growth on InN, which is expected to be in the tensile strain state, will also be discussed.



Fig. 1 One of RSM obtained in this study and evolution of peak position for (10-11) diffraction peak of GaInN as function of growth time.



Fig. 2 Evolution of In composition and relaxation ratio of GaInN during growth.



Fig. 3 Evolution of integrated intensity of GaInN (10-11) peak during growth.

## Acknowledgements

This work was partly supported by MEXT Nanotechnology platform and the synchrotron radiation experiments were performed at the BL11XU of SPring-8 (Proposal Nos. 2015A3512, 2016A3562 and 2017A3587). This work was also partly supported by JSPS KAKENHI Grant Numbers #15H03559 and #17H02778.

### References

- T. Yamaguchi, N. Uematsu, T. Araki, T. Honda, E. Yoon and Y. Nanishi, J. Cryst. Growth **377**, 123 (2013).
- [2] K. Hiramatsu, Y. Kawaguchi, M. Shimizu, N. Sawaki, T. Zheleva, R. F. Davis, H. Tsuda, W. Taki, N. Kuwano and K.

Oki, MRS Internet J. Nitride Semicond. Res. 2, 6 (1997).