Synchrotron X-ray Scattering Study of thin AlN /Sapphire(0001) Films

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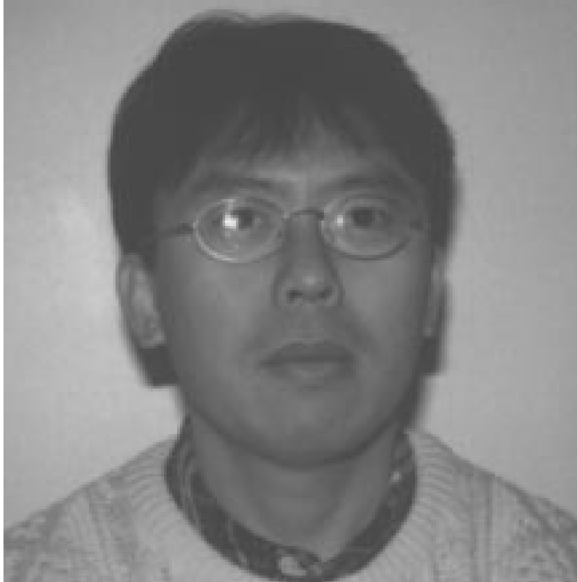

Editors' Note: The author has received his Ph.D from MIT with X-ray Scattering Studies. He has been a staff scientist at Exxon Research Engineering Company and He is currently a professor of Materials Science and Engineering at Kwangju Institute of Science and Technology. Professor Noh's e-mail address is "dynoh@kjist.ac.kr".

I. Introduction

Recent advances in blue light emitting materials have attracted much attention to III-nitride semiconductors [1]. One of the major difficulties in fabricating high quality nitride films such as AlN and GaN has been the absence of lattice-matched substrates [2]. GaN films are grown mostly on sapphire (0001) substrates which have the lattice mismatch as large as 16.1 %. The large lattice mismatch causes very rough and structurally disordered initial GaN layers. On the other hand, in case of AlN, two-dimensional growth of high quality layers on sapphire has been reported, albeit with the large lattice mismatch of 13.3 % [3,4]. Two-dimensional AlN is a potential candidate for the substrate to grow high quality GaN on. Understanding the strain evolution of the AlN is then a significant issue in the field of nitride semiconductor films.

The planar growth of AlN was explained by the extended domain matching (EDM) model wherein eight Al distances in AlN match to nine Al distances in sapphire [5]. The residual misfit then becomes 0.7 % which is small enough to allow two-dimensional planar growth. In the EDM model, most of the strain is relaxed by generating edge type dislocations at every eight Al atoms of AlN in the very early stage growth. The dislocations, which provide nearly commensurate condition, make the initial thin AlN layer fully strained. As film thickness increases, the strain would be relaxed to reduce the accumulated strain energy by generating additional misfit dislocations or undulating the growth front [6,7]. The undulation of the growing surface might provide nucleation sites for three-dimensional islands. It has been reported that most AlN films show the typical three-dimensional columnar growth in the late stage [6,8].

In this paper, we present a high-resolution synchrotron x-ray scattering study on the strain evolution of AlN/sapphire(0001) thin films. By investigating AlN films of various thickness, we reveal the detailed conditions of EDM in thin films as well as the nature of the strain evolution. The highly asymmetric scattering profile of thick AlN films was attributed to the strain distribution across the film.

II. Experimental Setup

The AlN films were grown on single crystal sapphire (0001) substrates in a radio frequency (RF) magnetron sputtering deposition system. A pure Al (99.999 %) target of about 50 mm in diameter was used, and the sapphire substrates was placed about 40 mm away from the target. As the sputtering gas, 5×10^{-3} Torr of pure N_2 (99.999 %) or Ar/N_2 mixture of various compositions was used. The RF power was varied from 30W to 200W. The data shown in this paper were obtained on the films grown under pure N2 gas and 50W of the sputtering power, since they illustrated the evolution of the strain most clearly. The growth rate was 25Å/min.

The synchrotron x-ray scattering measurements were performed at beamline 5C2 at Pohang Light Source in Korea. The x-ray beam was focused by a mirror, and monochromatized by a double bounce Si(111) monochromator to 1.45Å. The diffraction measurements were performed mainly around the AlN(0002) reflection along the substrate normal, and the AlN(10$\bar{1}$2) that has finite momentum transfer in the plane of the film.

III. Results and Discussions

Figure 1 shows the scattering profile around the AlN (0002) and the AlN(102) Bragg peaks in a 85Å thick AlN film. The longitudinal scan (Fig. 1(a)) of the (0002) reflection shows interference fringes along the film

normal, q_z direction, which is a typical signature of a structurally coherent finite size system. The thickness of the film was estimated from the period of the interference fringes. The transverse scan (Fig. 1(b)) of the (0002) reflection, equivalent to the theta rocking curve, shows an extremely sharp peak with negligible diffuse scattering. The half-width at the half maximum (HWHM) was only about 1×10^{-4}Å^{-1} (0.0025°), which indicates that the layered structure was correlated over 3μm in the film plane direction. Although the sharpness of the transverse scan does not guarantee the long-range in-plane order of the atomic position in a layer [9], it indicates that the crystalline mosaic was extremely small, and the interface wa

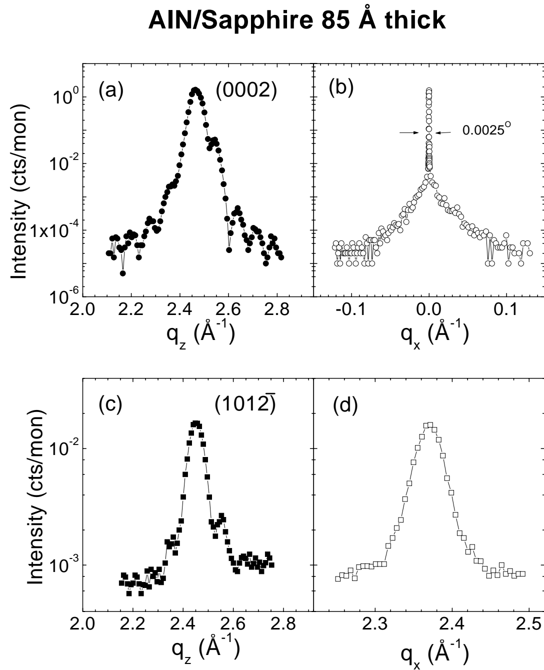

FIG. 1. Diffraction profiles of an 85Å thick AlN film. (a,b) AlN(0002) reflection in the surface normal and in the film plane directions respectively. (c,d) AlN(10$\bar{1}$2) reflection in the surface normal and the in the film plane directions respectively.

To investigate the structural properties in the film plane q_x direction, it is necessary to examine the (10$\bar{1}$2) reflection that has a non- vanishing momentum transfer component in the film plane as well as in the normal direction. The existence of the well-defined (10$\bar{1}$ 2) reflection illustrated in Fig. 1 (c,d) shows that the thin AlN film is grown epitaxially on sapphire in wurtzite structure. The diffraction profile along the surface normal direction illustrated in Fig. 1(c) shows interference fringes similar to those observed at the AlN(0002), although it is less pronounced. This suggests that the film be free of defects affecting the in-plane positional order of atoms along the film normal direction through out the whole film thickness. The structural coherence was only limited by the finite film thickness. In contrast to the sharp transverse profile of the (0002) peak, the (10$\bar{1}$2) peak, shown in Fig. 1(d), is quite broad in q_x direction with HWHM of 0.018Å^{-1}. The structural coherence length or the crystal domain size was about 180Å which was estimated roughly by π/HWHM. This length can be thought as the typical separation of the in-plane defects such as dislocations and stacking faults.

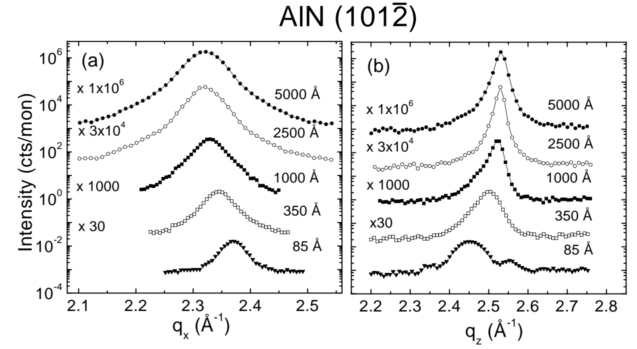

FIG. 2. Diffraction profiles of the AlN(102) in the film plane direction (a), and in the film normal direction (b)

The planar growth of epitaxial AlN thin films has been observed and explained by the extended domain matching (EDM) model.[5] In this model, edge dislocations generated at every eight AlN unit cells reduce the lattice strain immediately at the interface. Then eight unit cells of AlN (a = 3.112Å) grow on nine unit cells of sapphire (a =2.747Å) pseudo-morphically leaving only 0.7 % residual lattice strain. This amount of the lattice strain is small enough to allow the planar growth of AlN.
Our results on the 85Å thick AlN film exhibit that the detailed matching condition is slightly different from the EDM model. The in-plane lattice constant obtained from the peak position in Fig. 1(d) is 3.06Å. The in-plane strain is then about 2 % which is larger than 0.7 % estimated under the ideal 'eight-to- nine' matching condition. To explain the observed in-plane strain something close to 'ten-to-eleven' matching is required. Shim et al. also reported similar amount of strain on a thin AlN film grown by plasma-assisted molecular beam epitaxy [6]. The large initial strain indicates that less edge dislocations are generated at the interface than those required by the proposed EDM model. Presumably the high mobility of the depositing particles during the sputter growth made it less favorable to create edge dislocations at the interface. In fact, AlN films grown at lower RF power of 30W showed smaller strain of about 0.7 % as predicted by the EDM model. We might conclude that although the EDM model explains the planar growth of thin AlN films on sapphire, the exact matching condition is sensitive to the detailed growth conditions.

As the film thickness increases, the lattice strain becomes smaller progressively. Figure 2 shows the evolution of the AlN(102) peak obtained on AlN films of 85Å, 300Å, 1000Å, 2500Å, and 5000Å thick. Since the AlN(10$\bar{1}$2) reflection has both the in-plane and the out-of-

plane components of the momentum transfer, the lattice constants in respective directions can be readily obtained from the peak position through, $a=2\pi/q$. As the film thickness increases, the peak shifts toward higher q-values in the film normal direction, while it shifts to lower q-values in the film plane direction. This indicates that the in-plane lattice constant increases with increasing film thickness, while the out-of-plane lattice constant decreases. One might apply the Matthew-Blakeslee's argument [7] to understand the strain relaxation. As a film becomes thicker than the critical thickness, additional dislocations are generated to release the accumulated strain energy, which glide down to the interfacial region. The thicker the film, the more dislocations are generated. The thickness dependence of the strain could be explained in this model qualitatively.

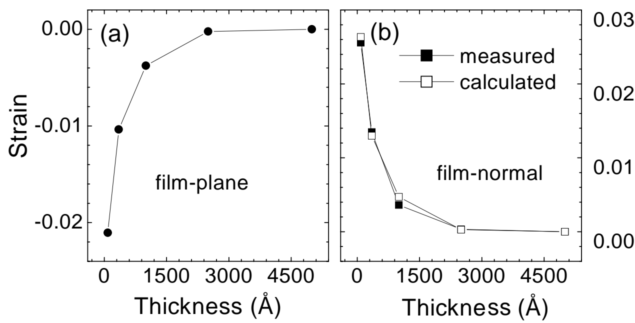

FIG. 3. (a) The lattice strain in the film-plane direction. (b) The lattice strain in the film-normal direction. The calculated lattice strain is also displayed

The evolution of the lattice strain is quite consistent with the simple elastic theory of biaxially strained systems predicting, $\varepsilon_{\perp} = -\varepsilon_{\parallel}\, 2\,\nu/(1-\nu)$ where $\varepsilon_{\perp}(\varepsilon_{\parallel})$ is the out-of-plane (in-plane) strain and ν is Poisson ratio [10]. Figure 3 illustrates the evolution of the strain obtained from the measured lattice constants as a function of the thickness. The out-of-plane strain calculated from the in-plane strain using the above relation and the reported Poisson ratio of AlN, ν of 0.34, agree quite well with the measured values as illustrated in Fig. 3 (b). The figure shows that most of the strain was released as the film thickness reached to 1000Å.

The diffraction profile of the AlN(0002) in a thick film is highly asymmetric and complicated. Figure 4 shows the profile of the AlN(0002) of a 2500Å thick film. The asymmetric shape in the longitudinal scan indicates that the lattice constant in the corresponding q_z direction varies significantly across the film. While the Matthew-Blakeslee's argument explains the thickness dependence of the lattice strain, it assumes that the strain is uniform across the whole thickness in a film with given thickness.

We attribute the cause of the strain distribution to the columnar grains nucleated on top of the planar layer of the film during the growth. The transverse scan shown in Fig. 4(b) consists of a sharp and a broad component. The sharp component originates from the planar 2-D layer, while the broad component represents the 3-D columnar grains. The details of the crossover in the growth mode from the initial 2-D planar growth to later 3-D columnar growth will be reported separately [11]. As the columnar grains start to grow, the 2-d layer underneath stops growing. Therefore the thickness of the 2- D layer would not increase any more once the columnar grains are nucleated. Since the columnar grains do not occupy the whole growing surface at once, but occupy the surface progressively, the thickness of the 2-D layer varies across the film plane. Since the amount of the strain relaxation depends on the film thickness as argued by Matthews and Blakeslee, the strain of the 2-D layered part would be non-uniform across the film.

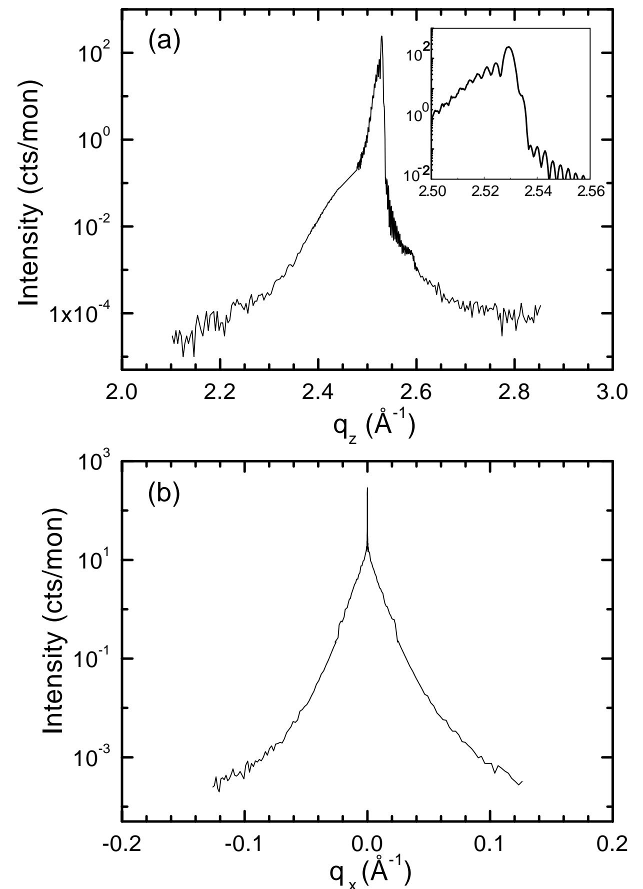

FIG. 4. (a) Longitudinal scan of AlN(0002) peak of a 2500 Å thick film. The inset illustrates the central part of the peak. (b) Transverse scan of the AlN(0002) peak.

The longitudinal profile shown in Fig. 4(a) mostly represents the sharp component of the transverse profile caused by the 2-D layer since it is taken at the peak of the transverse scan. The longitudinal profile consists largely of two components; a sharp central peak occurred near the bulk position of the AlN(0002) and a broad component at smaller q-side. The sharp central peak is attributed to the thick part of the 2-D layer where most of the lattice strain was released. Meanwhile the broad component at smaller

q-side represents the highly strained thinner part of the 2-D layer. We note the fine interference fringes near the central peak, which indicates that the thick part of the 2-D layer is structurally coherent across the whole thickness of the film.

IV. Summary

In summary, we studied the strain evolution of AlN/Sapphire (0001) films in a high-resolution synchrotron x-ray scattering experiment. The thinnest 85 Å thick film was a highly strained planar film. The domain matching condition was close to 'ten-to-eleven' different from the reported 'eight-to-nine' matching. With increasing the film thickness, the strain was relaxed. The relation between the in-plane and the out-of-plane strain was well described by the elastic theory of a biaxially strained system. In a thick film, the three-dimensional columnar grains that limit the thickness of the 2-D layer underneath impeded the strain relaxation. The thickness variation of the 2-D layered structure resulted in a non-uniform strain distribution. The asymmetric AlN(0002) reflection of thick AlN films was explained by this non-uniform strain relaxation.

References

[1] J. H. Edgar, Properties of Group III Nitrides, (INSPEC, Reading, UK) (1994).

[2] S. Nakamura, and G. Fasol Appl. Phys. Lett. **67,** (11), 1549 (1997).

[3] Daudin, and F. Widmann, J. Cryst. Growth. **182**, 1 (1997).

[4] R. D. Vispute, H. Wu, and J. Narayan, Appl. Rev. Lett. **67** (11), 1945 (1997).

[5] C. J. Sun, P. Kung, A. Saxler, H. Ohsato, K. Haritos, and M. Razeghi, J. Appl. Phys. **75** (8), 3964, (1994).

[6] K. H. Shim, J. Myoung, O. Gluschenkov, K. Kim, C. Kim, and I. K. Robinson, Jpn. J. Appl. Phys. **37** L313, (1998).

[7] J. W. Matthews and A. E. Blakeslee, J. Cryst. Growth **27** 118 (1978).

[8] M. Yeadon, M. T. Marshall, F. Hamdani, S. Pekin, H. Morkoc, and J. M. Gibson, J. Appl. Phys. **83** (5), 2847, (1998).

[9] P. R. Miceli, and C. J. Palmstrom, Phys. Rev. B **51** 5506, (1995).

[10] L. D. Landau and E. M. Lifshitz, Theory of Elasticity, 3rd. ed. (Pergamon, New York, 1986), p. 46.

[11] H. C. Kang, S. H. Seo, and D. Y. Noh, manuscript in preparation.