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Initial Growth Mode and Epitaxial Growth of AlN Thin Films on Al_2O_3(0001) Substrate by DC Faced Target Sputtering

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Using *DC* faced target sputtering method we grew AlN thin films on the Al_2O_3(0001) substrate with varying thickness(17 Å-1000 Å). We measured x-ray diffraction(XRD) profiles by synchrotron radiation(λ=1.12839 Å) with four circle diffractometer. The full width half maximum(FWHM) of rocking curve for the AlN(0002) diffraction of the film grown at 500 °C was 0.029 °. Also, we confirmed that the stress between AlN thin film and Al_2O_3(0001) substrate was reduced as increasing AlN film thickness, and the critical thickness of 400 ~ 500 Å, defined as a lattice constant in the film agrees with that in a bulk without stress, was obtained.

Key words: AlN, Critical Thickness, Epitaxy

I. Introduction

Recently AlN thin films are of interest due to the potential for several kinds of applications such as passivation and dielectric layers in electronic devices, short wavelength emitters, UV photonic detectors, and high-frequency band surface acoustic wave devices because of their high thermal conductivity, excellent piezoelectricity, high acoustic velocity, and wide band gap of 6.2 eV.[1,2] Moreover, the band gap is tunable from 6.2 to 3.4 eV or from 6.2 to 1.9 eV if the $Al_xGa_{1-x}N$ or $Al_xIn_{1-x}N$ is formed, respectively. Epitaxial growth of AlN films on sapphire have been obtained in several techniques such as chemical vapor deposition,[3,4] pulsed laser deposition,[5] atomic layer epitaxy.[6] But there are only a few reports on the high quality epitaxial growth of AlN films on sapphire at relatively low temperatures(less than 750 °C)[4,5] In this paper, we report epitaxial growth of (0001)AlN thin films on (0001)Al_2O_3 substrates at 500 by reactive dc faced magnetron sputtering method.

The deposited unit cell of AlN is located on that of sapphire substrates with a rotation of 30 degrees. In this case the lattice mismatch between thin film and substrate is more than 13%. This value is very large and epitaxial growth for such a large lattice mismatch system can be explained by domain matching epitaxy(DME)[5,7] or extended atomic distance mismatch(EADM)[8] model. This misfit is either accomodated entirely by strain or is shared between dislocation and strain.[9] In this study we observed the misfit accomodation of AlN films on sapphire by measuring lattice constant of films and the thickness of which the mismatch stress is completely relaxed.

II. Experimental Procedure

The AlN films were deposited on Al_2O_3(0001) substrate using the homemade *DC* faced target sputtering system. The two one-inch Al targets faced to each other with 5 cm distance. The substrates were cleaned ultrasonically with acetone and deionized water, and dried with nitrogen gas. The Al targets were presputtered for 10 minutes to remove oxides and impurities. Substrate temperatures were controlled within ±2 °C at 500 °C. The deposition time was varying with 8-420 sec and the deposition rate was 2.4-2.7 Å/sec. The growth processes have been described previously.[10]

The x-ray diffraction(XRD) profiles was measured using the beamline X-16C of the National Synchrotron Light Source (λ=1.12839 Å) with four circle diffractometer. To determine the lattice constants of AlN epitaxial layers, we measured XRD Bragg peaks for out of plane(0002) and in-plane($10\bar{1}1$) of AlN thin films. In this way, lattice constant *a* and *c* were obtained for each sample. Also, the thicknesses of AlN epilayer were calculated from the oscillation periods of XRD profiles of AlN(0002) diffraction for each samples.

III. Results and Discussion

Fig. 1 shows the x-ray diffraction patterns in the growth direction of the films. The sputtered time and grown temperature were 100 sec and 500 °C, respectively In XRD profiles, we observed fringe from AlN(0002) diffraction and this fringe means that the films are highly textured with [0001] of AlN aligned with [0001] of Al_2O_3. Using the rela-

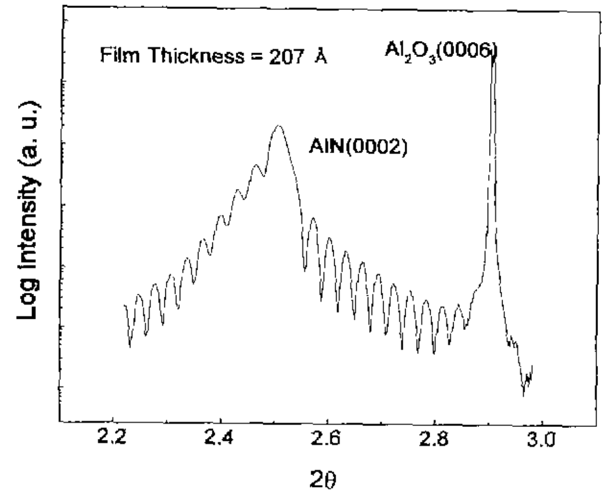

Fig. 1. XRD patterns of *DC* faced target sputtered AlN films on $Al_2O_3(0001)$.

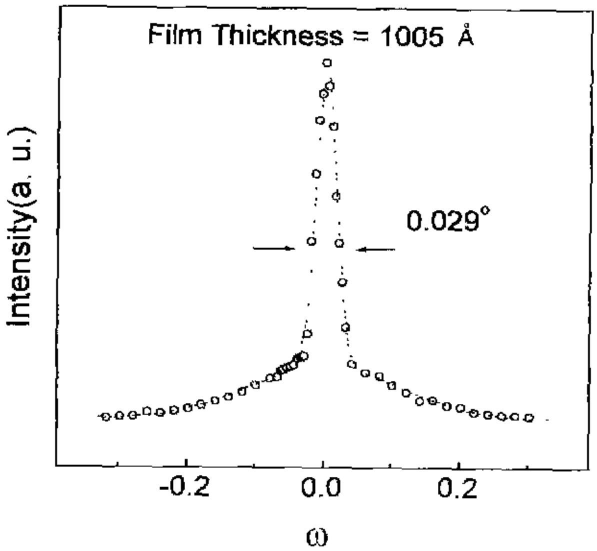

Fig. 2. Rocking curve for the AlN(0002) peak of the film grown at 500 .

tion for the film thickess, *Nd*, $Nd=2\pi/\Delta q$, where Δq ($q=4\pi sin\theta/\lambda$) is the momentum transfer difference between successive minima of fringes near the AlN(0002) Bragg peak, the AlN epilayer thickness of 207 Å was obtained. The thicknesses for each samples were calculated with same method.

The rocking curve for the AlN(0002) peak is shown in Fig. 2. The substrate temperature was 500 °C, and film thickness was calculated as 1005 Å. In Fig. 2, the FWHM of rocking curve was 0.029 ° which is the narrowest value reported to our knowledge grown less than 500 °C.

The FWHM *of rocking curve of* 0.029 ° *is* very close to that of instrumental resolution limit. It means that the AlN film is high quality of single crystal.

Fig. 3 shows azimuthal(φ) diffraction patterns for the internal planes of the AlN film and sapphire substrate. We can see the clear 6-diffraction peaks for internal planes of AlN epilayer It means AlN has hexagonal structure and grows in the epitaxial formation. Moreover the φ value of six XRD peaks for AlN$(10\bar{1}1)$ and $Al_2O_3(11\bar{2}3)$ planes are exactly same with each other. Similarly, Fig. 3 also shows same φ value of XRD peaks for AlN$(11\bar{2}2)$ with $Al_2O_3(10\bar{1}4)$ planes and also the difference of φ value between AlN$(11\bar{2}2)$ and $Al_2O_3(11\bar{2}3)$ planes are exactly 30 °. This result revealed that the in-plane relationships of AlN film with respect to the sapphire substrate in the basal *a-b* plane are 30 ° *rotation and these are identical with previously* reported results.[5,6] Furthermore, the in-plane epitaxial relationship were found to be AlN$(10\bar{1}0)$//$Al_2O_3(11\bar{2}0)$. In this case the Al-Al distance in the $[1\bar{1}0]$ direction of Al_2O_3 is 2.747 Å, and that of AlN in [100] is 3.112 Å. Thus lattice mismatch is 13.29 % and which is relatively large.

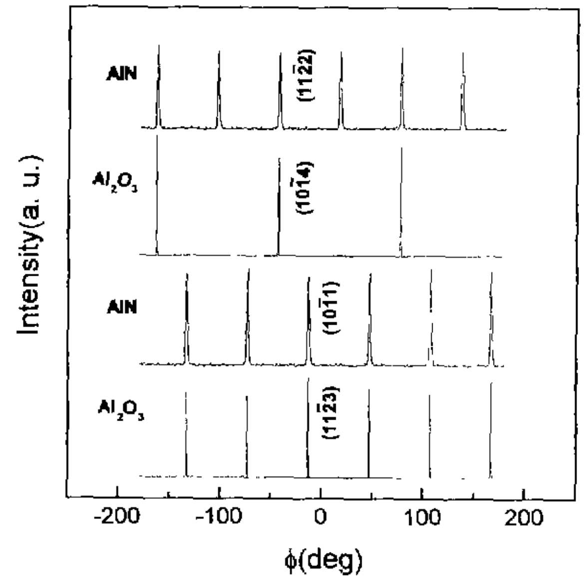

Fig. 3. Azimuthal (φ) diffraction patterns for internal-plane of AlN(0001)/$Al_2O_3(0001)$.

Fig. 4 shows lattice constant *a* and *c* as a function of AlN

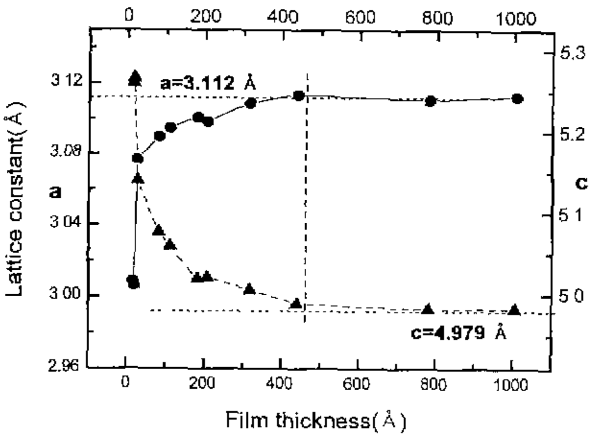

Fig. 4. Lattice constants *a and c as a function of AlN layer* thickness.

layer thickness. Caused by lattice misfit AlN layer gets compress-stress along an *a*-axis and lattice constant *a* of AlN layer is reduced. The compress-stress imposed in the parallel direction causes stretch-stress along the perpendicular direction. It gives increasing lattice constant along the c-axis. In early growth stage, the AlN layer thickness was calculated as about 17 Å from XRD fringes, and the lattice constant of *a* and *c* was obtained as 3.008 and 5.259 Å, respectively. Compare with stress-free AlN lattice constants, a=3.112 and c=4.979 Å, the AlN epilayer has largely strained because of lattice mismatch. To explain epitaxial growth for the large mismatch system, DME[5,7] and EADM[8] models were suggested. According to these model, lattice matching ratio of film and substrate is $m{:}n$, where m, n is integer but not always one. From the measurements of lattice constant of very thin AlN layer (about three-layer), a=3.008 Å was obtained, and in this case, one can say the matching ratio of AlN:Al_2O_3 is about 10:11 by DME model. Also, Fig. 4 shows progressive trend away from the bulk AlN lattice constants for thinner film. At thermodynamic equilibrium, misfit dislocations appear at the interface of strained layer hetrostructure when the strained is thick enough that it is energetically favorable for the mismatch to be accommodated by a combination of elastic strain and interfacial misfit dislocations, rather than by elastic strain alone.[11] According to the thickness of AlN layer increased the dislocation density increased and the strained layer was relaxed. When the AlN layer thickness was increased to 400-500 Å, the strain was fully relaxed and the lattice constants of layer converged to the bulk balue.

IV. Conclusion

In summary, the high quality epitaxial AlN thin films have been grown on basal plane sapphire substrates [(0001)Al_2O_3] at low temperatures(500 °C) by dc faced target sputtering method. The FWHM of rocking curve for the AlN(0002) peak of the AlN film was 0.029° which is the narrowest value reported grown less than 500 °C. From measuring x-ray diffraction, the stress between AlN thin film and Al_2O_3(0001) substrate is relaxed as increasing AlN film thickness and the critical thickness of 400~500 Å, defined that a lattice constant in the film agrees with that in a bulk without the stress, was obtained. Also, the matching ratio of AlN:Al_2O_3~10:11 was obtained.

References

1. P. K. Kuo, G. W. Auner and Z. L. Wu, "Microstructure and Thermal Conductivity of Epitaxial Aln Thin Films." *Thin Solid Films*, **253**, 223-227 (1994).
2. S. Strite and H. Morkoc, "GaN, AlN and InN: A Review," *J. Vac. Sci. Technol.*, **B10**(4), 1237-1266 (1992).
3. A. Saxler, P. Kung, C. J. Sun, E. Bigan and M. Razeghi, "High Quality Aluminum Nitride Epitaxial Layers Grown on Sapphire Substrates," *Appl. Phys. Lett.*, **64**(3), 339-341 (1994).
4. W. Zhang, Y. Someno, M. Sasaki and T. Hirai, "Low-Temperature Epitaxial Growth of AlN Films on Sapphire by Electron Cyclotron Resonance Plasma-assisted Chemical Vapor Deposition," *J. Crystal Glowth*, **130**, 308-312 (1993).
5. R. D Vispute, Hong Wu and J. Narayan, "High Quality Epitaxial Aluminum Nitride Layers on Sapphire by Pulsed Laser Deposition," *Appl. Phys. Lett.*, **67**(11), 1549-1551 (1995)
6. M. A. Khan, J. N. Kuznia, R. A. Skogman and D. T. Olson, "Low Pressure Metalorganic Chemical Vapor Deposition of AlN over Sapphire Substrates," *Appl. Phys Lett.*, **61**(21), 2539-2541 (1992).
7. T. Zheleva, K. Jangannadham and J Narayan, "Epitaxail Growth in Large-lattice-mismatch Systems," *J Appl. Phys.*, **75**(2), 860-871 (1994).
8. C. J. Sun, P Kung, A sacler and M. Razeghi, "A Crystallographic Model of (001) Aluminum Nitride Epitaxial Thin Film Growth on (001) Sapphire Substrate," *J. Appl Phys.*, **75**(8), 3964-3967 (1994).
9. J. W. Matthews, S. Mader and T B. Light, "Accommodation of Misfit Across the Interface Between Crystals of Semiconducting Elements or Compounds," *J. Appl. Phys.*, 41(9), 3800-3804 (1970).
10. J. W. Kim and H. K. Kim, "Low Temperature Epitaxial Growth of AlN Thin Films on Sapphire by Reactive dc Faced Magnetron Sputtering," *J. Kor. Phys. Soc.*, **32**, S1664-S1666 (1998).
11. J. Kang and T. Ogawa, "Misfit Dislocations and Stresses in GaN Epilayers," *Appl. Phys. Lett.*, **71**(16), 2304-2306 (1997).