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Radio-frequency MBE growth of cubic GaN on 3C-SiC(001)/Si(001) template

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A 3C-SiC(001) was formed by coincidence site lattice matching on Si(001) through carbonization using acetylene (C_2H_2) supplied by a jet nozzle. The 3C-SiC serves as a cubic template for the MBE growth of c-GaN on Si substrates. The Si substrate with lattice constant of a_{Si} = 0.544 nm was covered with strained 3C-SiC whose lattice constant $a_{3C\text{-}SiC/Si}$ = 0.445 nm expanded from that of the bulk 3C-SiC crystal (0.438 nm). Better quality cubic phase GaN film was grown on the strained 3C-SiC template layer when the Ga/N flux ratio approached unity of Ga rich side. The proportion of the cubic phase in the GaN layer was calculated to be 94.7% from the integrated intensity ratio of the X-ray diffraction peaks of c-GaN(002) and h-GaN(10-11). The FWHM of the (002) diffraction peak for the 544 nm-thick c-GaN was 13.2 arcmin. A precise RHEED analysis also indicated that the c-GaN had (2x2) surface reconstruction structure.

1 Introduction

Cubic gallium nitride, e.g. c-GaN, is more attractive than the hexagonal phase, h-GaN, since it has higher electron mobility, smooth cleaved surface and relative ease of p-type doping. Epitaxial growth of the metastable c-GaN requires a lattice-matched cubic substrate. The lack of homoepitaxial substrates for c-GaN forced heteroepitaxial growth techniques to be used. The coincidence site lattice (CSL) matching method [1], in which a coincidence site is defined as an atomic site that is common to both lattices. It is able to match different crystals by introducing misfit dislocations when their lattice parameter ratio is a special value such as 1/2, $\sqrt{2}$, $1/\sqrt{2}$,4/5, 5/6 *etc.* The ratios of the lattice parameter for 3C-SiC and c-GaN against Si are 4/5 and 5/6, respectively. The 3C-SiC/Si template therefore reduces the lattice mismatching with c-GaN and Si. Reduction of the anti-phase domain (APD) of c-GaN(001) is an important concern that needs to be addressed to obtain high crystal quality suited for applications in electronic and optical devices. The growth of c-GaN on SiN_x/Si substrate using microwave (μ) and radio frequency (RF) nitrogen discharges for MBE [2] and on 3C-SiC/Si template formed by carbonization of Si [3] have been previously reported. The RF-MBE method was widely studied because of the possibility of low growth temperature for c-GaN growth. Good quality c-GaN was achieved by the elimination of charged particles effusing from the nitrogen discharge [4, 5]. Review articles and monographs on RF-MBE growth were published in various forms [6–8]. In this report, a strained 3C-SiC template was formed by carbonization of Si substrate and successful growth of c-GaN(001) on the template by RF-MBE is presented.

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2 Experimental

A VG80-H MBE system equipped with an IRFS-501 RF nitrogen radical source made by Arios, Inc. was used to grow the c-GaN samples. In Fig. 1, a stainless-steel electrostatic cylindrical electrode was placed just after the orifice cap of the RF-plasma source to eliminate the flux of charged particles. The electrostatic electrode can be supplied with a DC-bias of negative to positive 1000 V to attain positive ions and electrons elimination from the nitrogen beam. An electromagnet was also installed to deflect mainly electrons as shown in Fig. 1. To detect excited neutral nitrogen species a Pt electrode as a atomic probe was placed near the surface of substrate as shown in Fig. 1. Two Ga K-cells were used to low temperature buffer (LTB) for small Ga flux and c-GaN growth.

Prior to the surface carbonization of a 2 inch or 3 inch Si(001) substrate, it was chemically treated by the Ishizaka and Shiraki method [9] with boiling HNO_3, NH_4OH and HCl, and was successively rinsed with deionized water. The 3C-SiC/Si(001) templates were obtained through Si substrate carbonization in ultra high vacuum. The 3C-SiC was grown by alternating exposure of Si flux supplied by a K-cell and acetylene (C_2H_2) flux from a jet nozzle [3]. Carbonization time was for 15 min at 570 °C after formation of amorphous SiC at 440 °C. Complete carbonization and good quality 3C-SiC template was confirmed using RHEED. A digitizer software, Imm Core (Version1.3.0.28) of Hamamatsu Photonics K.K. was used to convert from an RHEED pattern to digital data. The RF power and the nitrogen flow rate were kept at 500 W and 0.5 sccm, respectively to maintain unity III/V ratio of 300 nm/h at the growth surface. Migration enhanced epitaxy (MEE) [10] was also effective in suppressing the formation of Ga droplets on the strained 3C-SiC template. Cubic-GaN(001) was only obtained under uniform N* and N_2* flux distribution and at stoichiometric or at slightly Ga rich condition. The crystalline structural purity of c-GaN (001) was calculated from the ratio of the integrated peak intensities between cubic (002) peak and total intensity including hexagonal peak. Crystal quality of c-GaN(001) was confirmed through observation of the reflection high energy electron diffraction(RHEED) and atomic force microscopy, X-ray diffraction, cross-sectional high resolution transmission electron microscopy(HRTEM), and photoluminescence measurements.

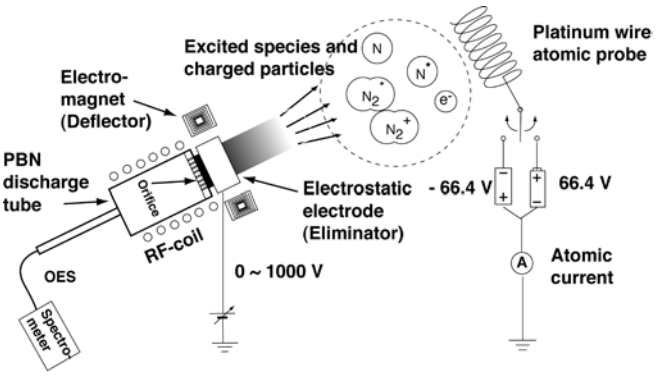
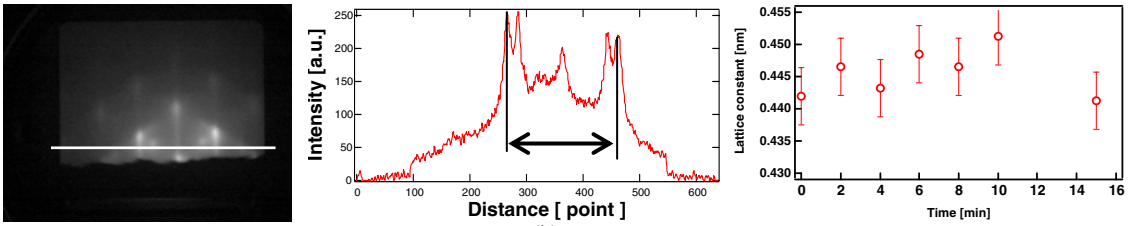


Fig. 1 Schematic diagram of a nitrogen radical cell, charged particle eliminator and deflector, and the activated neutral species flux measurement set-up of Pt electrode.

3 Results

Digital data of the intensity profile of RHEED pattern on the white line in Fig. 2(a) is given in Fig. 2(b). Distorted lateral lattice constant of the 3C-SiC on Si was calibrated using the Si streak spacing with the standard temperature dependence lattice constant of a_{Si}= 0.5441 nm at 570 °C [11]. Figure 2(c) shows the change of the 3C-SiC lattice constant measured from the peak distance of a RHEED pattern during the carbonization of Si for 15 min at 570 °C. The averaged lattice constant is $a_{3C\text{-}SiC/Si}$ = 0.445 nm from Fig. 2(c). The peak positions, which were obtained from the digitized data, contained error of 1.6 % originated from the resolution of digitized data. It was found that the 3C-SiC template had a larger lateral lattice constant than that of a bulk 3C-SiC, a = 0.4376 nm at 604 ^{o}C [12]. The strained lateral lattice constant $a_{3C\text{-}SiC/Si}$ = 0.445 nm could be suitably matched to the lattice constant of c-GaN (a = 0.4504 nm at room temperature) [13].



(a) (b) (c)

Fig. 2 The expansion of 3C-SiC lattice constant measured from the RHEED pattern peak distance. (a) RHEED pattern observed from <110>. (b) The intensity profile of RHEED pattern on the white line of Fig. 2(a). (c) The trend of the lattice constant change during the annealing at 570 °C for the carbonization of Si by C_2H_2

Figure 3 shows HRTEM observation of a grown c-GaN/3C-SiC/Si(001) with the interface of LTB from [1-10] orientation. The interface between 3C-SiC and Si shows lattice matching by CSL introducing misfit dislocations. The white contrast of Si near the SiC/Si interface shows the possibility of the compression of Si lattice. The interface between c-GaN and 3C-SiC also shows the existence of misfit dislocations. The thickness of 3C-SiC layer was about 2.5 nm as shown in Fig. 3.

A LTB GaN was grown at 430 °C using MEE method to ensure a smooth and uniform growth surface. The change of RHEED patterns of c-GaN on 3C-SiC/Si (001) caused by the varying Ga/N flux ratio is shown in Fig. 4. Under slightly nitrogen excess condition; island formation was recognized from spotty patterns and additional spots indicate h-GaN inclusions, as shown in Fig. 4(a). Streaky pattern under unity Ga/N ratio or slightly Ga excess condition of c-GaN (001) is observed in Fig. 4(b). Under Ga excess condition, the faint spotty pattern in Fig. 4(c) confirms the formation of Ga droplets.

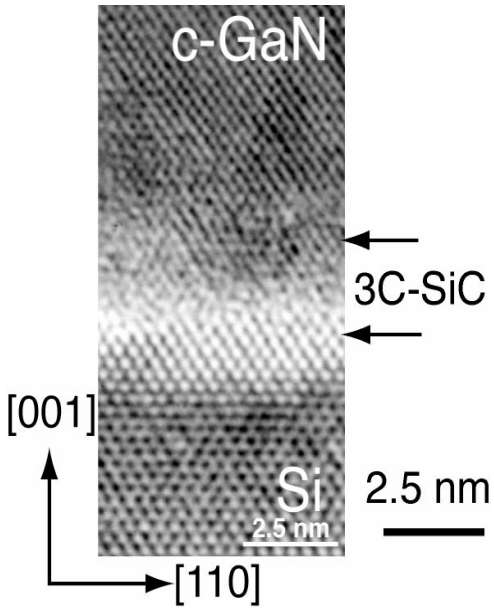
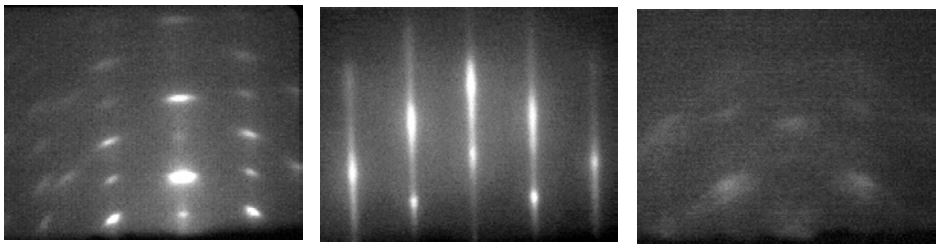


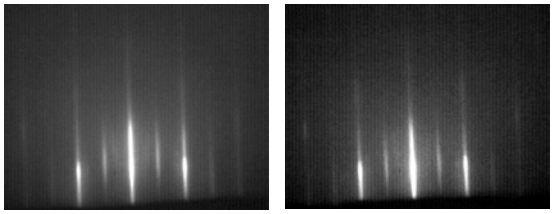
Fig. 3 Cross-sectional TEM observation of the interface of c-GaN(001)/3C-SiC(001)/Si(001).



(a) (b) (c)

Fig. 4 RHEED patterns of c-GaN on 3C-SiC/Si (001) under different Ga/N flux ratio; (a) under slightly nitrogen excess condition; island formation was obvious from spotty patterns and additional spots indicate h-GaN inclusions, (b) under unity Ga/N ratio of slightly Ga excess condition; weak spotty pattern is derived from c-GaN (001), (c) under slightly Ga excess condition; Ga deposition causes weaken the spot pattern was confirmed. The direction of electron beam is parallel to [-110].

The bulk GaN was grown at 750 ^{o}C for 2 hrs at Ga/N flux ratio approaching unity. A predominantly cubic phase GaN film was observed to grow on the strained 3C-SiC template layer. The proportion of cubic phase in the GaN layer was calculated to be 94.7% from the ratio of the integrated intensity of c-GaN(002) and hexagonal-GaN(10-11) X-ray diffraction peaks. In addition to this, the FWHM of the (002) diffraction peak for the 544 nm-thick c-GaN was 13.2 arcmin. A precise RHEED analysis also indicated that the c-GaN had (2x2) surface reconstruction structure as shown in Fig. 5.



(a) *e* || [110] (b) *e* || [-110]

Fig. 5 RHEED patterns taken from different incident beam directions, [110] (a) and [-110] (b) for a surface of 2.5 μm thick c-GaN grown under slightly Ga excess condition. The (2x2) reconstruction was also observed due to the existance of double domains.

4 Conclusion

The expansion of the lattice constant of the 3C-SiC template was confirmed from the RHEED patterns during carbonization of the Si substrate. The strained lattice constant was favorable for cubic GaN growth. Successful growth of c-GaN was verified by XRD analysis.

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References

[1] W. Bollmann, Surf. Sci. **31,** 1 (1972).
[2] T. Ohachi, T. Kikuchi, Y. Ito, R. Takagi, M. Hogiri, K. Miyauchi, M. Wada, Y. Ohnishi, and K. Fujita, phys. stat. sol. (c) **0**(7), 2589 (2003).
[3] T. Kikuchi, K. Miyauchi, M. Wada, and T. Ohachi, J. Cryst. Growth **275(1 – 2)**, e1215 (2005).
[4] T. Ohachi, T. Kikuchi, K. Miyauchi, Y. Ito, R. Takagi, M. Hogiri, K. Fujita, O. Ariyada, and M. Wada, J. Cryst. Growth **275,** e1197 (2005).
[5] A. Somintac, T. Kikuchi, N. Fujii, K. Shibataki, O. Ariyada, T. Ohachi, and M. Wada, phys. stat. sol. (c), to be published.
[6] A. Georgakilas, H. M. Ng, and P. Komninou, in: Nitride Semiconductors, Handbook on Materials and Devices, edited by P. Ruterana, M. Albrecht, and J. Neugebauer (Wiley-VCH, New York/Weinheim, 2003), p. 107.
[7] H. Morkoç, S. Strite, G. B. Gao, M. E. Lin, B. Sverdlov, and M. Bums, J. Appl. Phys. **76,** 1363 (1994).
[8] B. Schottker, Molecular Beam Epitaxy and Characterization of Doped and Undoped Cubic GaN Layers (Shaker Verlag, Aachen, 1999); ISBN 3-8265-6626-2.
[9] A. Ishizaka and Y. Shiraki, J. Electrochem. Soc. **133,** 666 (1986).
[10] Y. Horikoshi, M. Kawashima, and H. Yamaguchi, Jpn. J. Appl. Phys. **27**, 169 (1988).
[11] W. M. Yim and R. J. Paff, J. Appl. Phys. **45**, 1456 (1974).
[12] G. A. Slack and S. F. Bartram. J. Appl. Phys. **46,** 89 (1975).
[13] X. H. Zheng, Y. T. Wang, Z. H. Feng, H. Yang, H. Chen, J. M. Zhou, and J. W. Liang, J. Cryst. Growth **250**, 345 (2003).

