

High quality AlGa_N growth by changing growth pressure and insertion of AlN/GaN superlattice interlayer

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We have investigated the optical and structural properties of thick AlGa_N layer grown using strain-relief AlN/GaN superlattice buffer on SiC/Si(111) substrate by low pressure metal organic chemical vapor deposition. A set of five period AlN/GaN superlattice interlayers was one of the most efficient methods for the strain relief by reducing biaxial tensile strain between GaN and AlGa_N layer. A set of five period AlN/GaN superlattice interlayer and a thick AlGa_N layer were grown by changing the growth pressure from 80 mbar to 100 mbar. The quality of AlGa_N layer strongly depends on the pressure of growth of the AlN/GaN superlattice interlayer and AlGa_N layer. The optimized pressure for the AlN/GaN superlattice interlayer and thick AlGa_N layer was 80 mbar and 100 mbar, respectively. Crystal quality and morphology of AlGa_N films improved and crack density reduced by using two-step growth method, remarkably.

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1 Introduction Wide band gap group of III-nitrides have been recognized as important materials for the optoelectronic devices, such as blue and ultraviolet (UV) light emitting diodes (LEDs), laser diodes (LDs), and high temperature/high frequency transistors [1, 2]. Currently intense researches are under way for developing deep UV optoelectronic devices for the application in solid-state white light, super high-density storage. Because of their direct band gap from 3.4 to 6.2 eV, high Al mole fraction AlGa_N layers are one of the most promising material for such devices. However, due to the biaxial tensile strain, crack network invariably generates when the thickness of the AlGa_N layer on Si substrate or GaN exceeds the critical value [3]. Therefore the production of crack-free thick AlGa_N ternary layer has been challenging issue due to the large lattice-mismatch (2.16%) between GaN and AlN. A considerable effort has been made to overcome the cracking problem by using AlGa_N/GaN superlattice [4], and by exploiting thin InGa_N stress relief layer beneath AlGa_N layer [3]. Recently, Amano et al. [5] reported on the growth of crack-free thick AlGa_N film by inserting a low-temperature (500 °C) grown thin AlN interlayer between GaN and AlGa_N layer. Han et al. reported on control of cracking of AlGa_N using a low-temperature AlGa_N interlayer [6, 7]. Also, thin high temperature (HT)-AlN also significantly reduces the tensile stress in the AlGa_N layer and is useful to grow crack-free high quality AlGa_N films [8]. Also, the growth of AlGa_N/GaN on Si substrate is particular interested. These methods can take advantage of the well-established Si technologies. Moreover, the availability of large and high quality Si substrate, as well as its low cost, make it an attractive alternative for the growth of III-nitride layers. In previous work we got the high quality GaN film on Si with AlN buffer using SiC intermediate layer because the SiC intermediate layer acts on reduce the lattice mismatch and reduces thermal expansion coefficient between GaN film and Si substrate [9].

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In this work, we investigated the growth of thick AlGaIn layer on GaN using strain-relief AlN/GaN superlattice (SLs) interlayer on SiC/Si(111) substrate by LP-MOCVD. We found that optimum conditions for the growth of AlGaIn/GaN layers strongly depended on the changing of the growth pressure of AlN/GaN SLs interlayer and thick AlGaIn layers. A set of five period AlN/GaN SLs and AlGaIn layer grown at 80 mbar and 100 mbar respectively improved the crystal and optical quality.

2 Experimental procedure Before growth of AlGaIn on GaN, the GaN film were grown with high temperature AlN buffer and five period AlN/GaN SLs buffer on Si(111) substrate using 3C-SiC intermediate layer. Si(111) substrate were thermally cleaned at 1100 °C for 5 min in H₂ ambient. Then the 3C-SiC intermediate layer was grown at 1250 °C and 50 torr by chemical vapor deposition (CVD) using tetramethylsilane (TMS). SiC/Si substrate was cleaned again in organic solvents and etched using BOE and boling HCl for 2 min, respectively and then flushed with deionized water. GaN films were grown at 1170 °C by LP-MOCVD using TMGa, TMAI, and NH₃ as Ga, Al, and N precursors, respectively. Before the growth of 20nm thick AlN buffer layer, the substrate was thermally treated at 1170 °C for 10 min in H₂ ambient. Then Al is directly deposited on the SiC/Si(111) substrate before introducing NH₃ into the reactor. The Al pre-seeding for 20 sec prevents the formation of Si–N bonding at the surface of SiC [9]. The thin AlN buffer layer was grown for 5 min with TMAI flow rate of 33 µmol/min and NH₃ flow rate of 1400 sccm at 100 mbar, respectively. After then five period AlN(2 nm)/GaN(5nm) SLs layer were grown for 15 sec and 30 sec, respectively. GaN epilayer on the AlN/GaN SLs buffer layer was grown for 1 hour with TMGa flow rate of 90 µmol/min and NH₃ flow rate of 2800 sccm at 1170 °C and 100 mbar. AlGaIn was deposited for 30 min on GaN epilayer using strain-relief AlN/GaN SLs interlayer with TMAI flow rate of 20 µmol/min, TMGa flow rate of 90 µmol/min and NH₃ flow rate of 3500 sccm at 1170 °C. The growth time of AlN/GaN SLs interlayer was 15 sec, 30 sec, respectively and the period of AlN/GaN superlattice was five period. The pressure of thick AlGaIn film and AlN/GaN SLs interlalyer was varied from 80 mbar to 100 mbar.

The surface morphology of AlGaIn films was studied using scanning electron microscopy (SEM) and an atomic force microscope (AFM). The Al solid composition and crystal quality of AlGaIn flims were measured by X-ray diffraction (XRD) and photoluminescence (PL) measurements. The thickness of AlGaIn layers on GaN film was observed using the field effect scanning electron microscopy (FESEM).

Using the transmission electron microscopy (TEM), we know reduction of dislocations of AlGaIn layer by preventing the dislocation of GaN epilayer.

3 Results and discussion As seen from Table 1, thick AlGaIn layers were grown by changing pressure of AlN/GaN SLs interlayer and thick AlGaIn flims. Sample A has no AlN/GaN SLs interlayer between GaN and AlGaIn layer, and sample B, C, D have a set of five period AlN/GaN SLs interlayer grown at different reactor pressure.

Table 1 Growth pressures for thick AlGaIn layers and AlN/GaN SLs of samples.

	A	B	C	D
Growth pressure of AlN/GaN SLs interlayer between AlGaIn and GaN	No	80mbar	100mbar	80mbar
Growth pressure of AlGaIn layers	80mbar	80mbar	100mbar	100mbar
solid composition of Al	12%	13%	10%	10%

Figure 1 shows SEM images of AlGaIn layers on GaN on the SiC/Si(111) substrate, where the thickness of AlGaIn layers is about 0.8 µm. Sample A which has no AlN/GaN SLs interlayer is many network of cracks and voids in the thick AlGaIn layer. These cracks were usually observed when the thickness of the AlGaIn layer exceeds 0.1µm. Cracks are caused by excessive biaxial tensile stress by activating slip system around the crack tips when the AlGaIn layer is over the critical thickness. We consider that the reduction of tensile stress can overcome the cracking problem through the insertion of strain-relief SLs

interlayers between AlGaN and GaN. In the sample B,C,D with the insertion of the AlN/GaN SLs interlayer between AlGaN and GaN layer, the tensile strain is alleviated [Figs. 1 (b),(c),(d)].

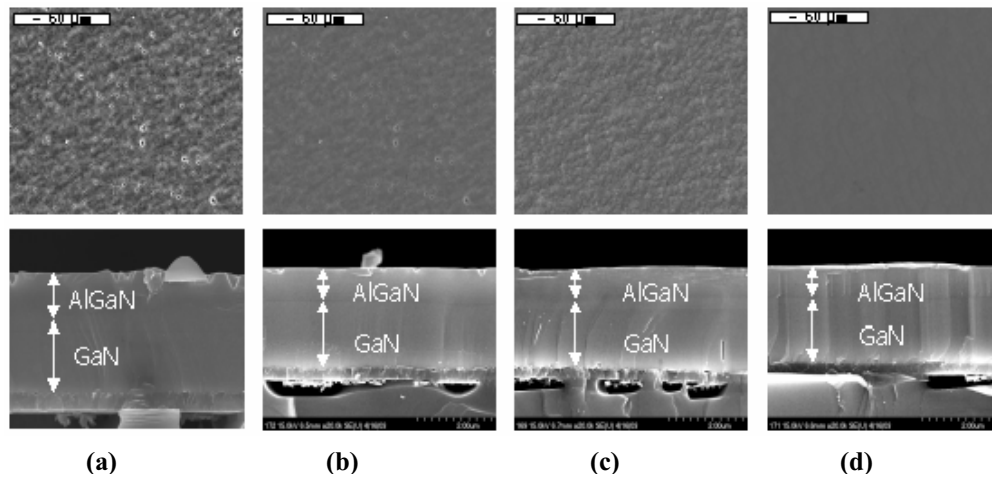


Fig. 1 SEM images of AlGaN layers grown on GaN at the conditions given in Table 1. (a) Sample A, (b) sample B, (c) sample C, (d) sample D.

As shown in Fig. 2, surface morphologies of AlGaN films were revealed by AFM investigations. The values of the root mean square (rms) of surface roughness are (a) 10.712 nm, (b) 2.716 nm, (c) 4.206 nm, and (d) 2.196 nm, respectively. As inserting of AlN/GaN SLs interlayer between AlGaN and GaN, surface morphology seems to be improved [Fig.2 (b), (c), (d)] corresponding to the SEM images. From these result, AlN/GaN SLs interlayer between AlGaN and GaN layers significantly reduced cracks and voids in AlGaN layers through the tensile strain alleviation.

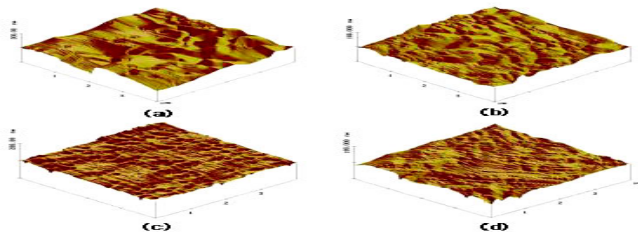


Fig. 2 AFM images of AlGaN layers grown on GaN at the conditions shown in Table 1. (a) Sample A, (b) sample B, (c) sample C, (d) sample D.

Generally, crystal quality and surface morphology strongly depend on reactor pressure. In case of single step growth, when AlN/GaN SLs and AlGaN layer were grown at same low pressure around 80 mbar, surface morphology was improved but crystal quality was deteriorated [Fig. 2 (b), Fig. 3(b)]. As increasing in reactor pressure to 100 mbar, crystal quality was improved but surface became rougher [Fig. 2(c), Fig. 3(c)]. In previous work, we studied growth of GaN layer by two-step growth [10]. In this work, we tried also to grow AlGaN layer by two-step method where AlN/GaN SLs interlayer and AlGaN layers have different reactor pressure to promote both crystal quality and surface morphology. Pressure of AlN/GaN SLs interlayer and AlGaN layer was varied from 80 mbar to 100 mbar. From the result, growth pressure of SLs interlayer was optimized at 80 mbar and thick AlGaN layer at 100 mbar [Figs. 1, 2, 3 (d)]. Crystal quality and morphology of AlGaN epi-layer improved and crack density reduced by using two-step growth method, remarkably. It is considered that the surface diffusion at low pressure and lateral growth at initial state dominate on the surface morphology of AlGaN epi-layer. The lateral growth of the first AlN/GaN SLs interlayers enhances surface morphology and reduces the strain in AlGaN

layer. As increase in pressure of first AlN/GaN SLs interlayers to 100 mbar, surface morphology is rougher and crack density is increased as shown in Fig. 1(c), Fig. 2(c). In this case, the higher vertical growth rate compared with the lateral growth rate makes discontinuous islands which are caused by many terraces and steps. It results in rough surface and high crack density in AlGaIn layer. Then the growth of discontinuous island is fast, and second AlGaIn layer is not fully coalesced. The root mean square (RMS) roughness of surfaces is reduced from 4.206 nm [Fig. 2(c)] to 2.196 nm [Fig. 2(d)] as decreasing pressure of first AlN/GaN SLs interlayer. Sample D has a good surface morphology in AlGaIn layer by lateral growth of first AlN/GaN SLs interlayer.

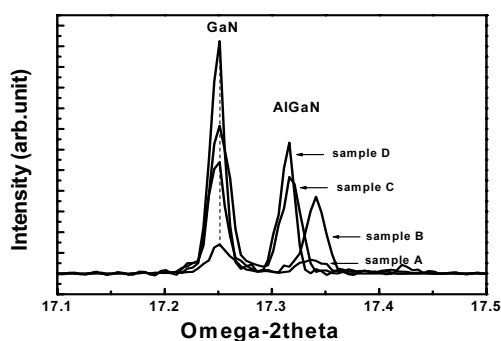


Fig. 3 HRXRD (2θ - ω) spectra of AlGaIn layers grown on GaN at the conditions given in Table 1. (a) Sample A, (b) sample B, (c) sample C, (d) sample D.

Figure 3 shows (0002) 2θ - ω XRD results. These results show that AlGaIn layers were well grown on GaN epi-layer. The Al-solid compositions of these samples were estimated from X-ray diffraction (XRD) and the photoluminescence (PL) results [Table 1]. The FWHMs of AlGaIn(0002) planes are (a) 188.04 arcsec, (b) 162.98 arcsec, (c) 149.37 arcsec, (d) 142.46 arcsec, respectively. Sample A has poor crystal quality due to tensile stress between AlGaIn and GaN. Because AlGaIn layer was grown without AlN/GaN SLs interlayer. In this sample the network of cracks and voids exists in AlGaIn layer [Fig. 1 (a)]. The other side, sample D has good quality of AlGaIn layer because AlN/GaN SLs interlayer and AlGaIn layer were grown by two-step method of changing the reactor pressure.

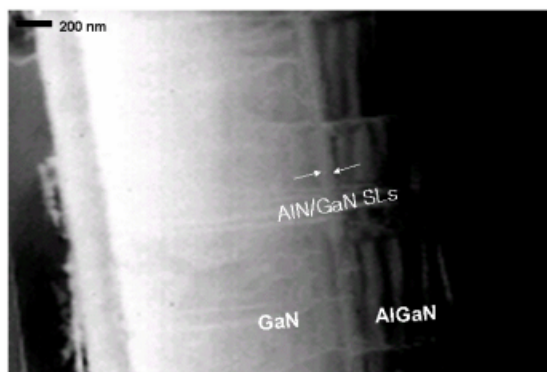


Fig. 4 TEM image of AlGaIn layers grown on GaN under sample D condition.

Figure 4 shows the transmission electron microscopy (TEM) image of AlGaIn flim grown on the GaN epi-layer with five period AlN/GaN SLs interlayer at same condition for sample D. As shown in Fig. 4, we obtained reduction of dislocation from GaN epi-layer using AlN/GaN SLs interlayer between GaN and AlGaIn layer.

4 Conclusions We have successfully grown thick AlGaIn layer on GaN using strain-relief AlN/GaN superlattice interlayer on SiC/Si (111) substrate by LP-MOCVD. We have investigated the properties AlGaIn layers by changing the growth pressure of AlN/GaN SLs interlayer and thick AlGaIn layers.

AlGaIn layer with AlN/GaN SLs interlayer is much better than AlGaIn layer without AlN/GaN SLs interlayer in structural properties and crystal quality. Especially, surface morphology and crystal quality was improved by two-step growth. The pressure of AlN/GaN SLs interlayer was optimised at 80 mbar and thick AlGaIn at 100 mbar. Then, we obtained the FWHM of XRD rocking curves of a AlGaIn(0002) plane was 142.46 arcsec, and the RMS value of surface roughness was 2.196 nm. We show also improved surface morphology and low dislocation density of AlGaIn layer through the images of SEM and TEM, respectively.

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