Achievement of Polarity Reversion From Al(Ga)-Polar to N-Polar For AlGa_N Film on AlN Seeding Layer Grown By a Novel Flow-Modulation Technology

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Achievement of polarity reversion from Al(Ga)-polar to N-polar for AlGa_N film on AlN seeding layer grown by a novel flow-modulation technology

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Abstract

N-polar AlGa_N epi-layer was realized on AlN seeding layer grown with a novel flow-modulation method. The polarity reversion from Al(Ga)-polar to N-polar for AlGa_N/AlN films was confirmed by KOH etching and subsequent observation with optical microscope as well as high-resolution X-ray diffraction (HR-XRD) measurement. In particular, the dependence of crystalline quality and defect size in the N-polar AlGa_N epi-layers on the V/III ratio was investigated with HR-XRD and scanning electron microscopy (SEM). It was found that as the full width at half maximum (FWHM) value of the X-ray rocking curve (XRC) varied with the V/III ratio in a "W-shape" for the N-polar AlGa_N epi-layers, and a FWHM value as small as 450 arcsec was achieved for the sample grown with a V/III ratio of 988. Moreover, it was revealed by the SEM measurement that the maximum diagonal length of hexagonal cone on the surface of the N-polar AlGa_N epi-layers decreased sharply when a V/III ratio of 1,236 was used although the crystalline quality and the surface morphology of the N-polar AlGa_N epi-layers were not improved simultaneously. The peculiar migration of the group-III atoms on the surface of the N-polar AlGa_N epi-layer associated with the molar ratio of TMA/(TMA+TMG) was considered to be

responsible for this result.

Keywords: N-polar AlGaN, crystalline quality, polarity reversion, surface morphology

1. Introduction

High electron mobility transistors (HEMTs) based on group-III nitrides such as GaN have attracted extensive and intense researches owing to their superior promise in breakdown voltage, electron saturation rate, intrinsic mobility, and thermal conductivity [1, 2]. It is known that the AlGaN/GaN heterojunction in a HEMT is usually fabricated to generate two-dimensional electron gas (2DEG) by utilizing the piezoelectric and spontaneous polarization-induced electrical fields [3]. Most of the schemes demonstrated to achieve enhanced HEMT so far include p-type gates, groove gates, F-ion processing, and Cascode structures generally consisted of Ga-polar nitrides [4-7]. Recently, the N-polar III-nitrides have become excellent candidates to implement high power, high frequency, and low loss HEMT application. The N-polar heterostructures possess a natural AlGaN back-barrier which can perform well in electron confinement, and thus reduce the short channel effect usually observed in the Ga-polar GaN-based HEMT [8].

In general, there are more and larger defects on the surface of the N-polar AlGaN film than on the surface of the Ga(Al)-polar AlGaN film which is usually used as the barrier layer in a HEMT. These defects will cause interface roughness-related scattering, thereby limiting the mobility of 2DEG [9]. In addition, the rough hexagonal hillock-like defects on the surface induced by the poor migration capability of the atoms will degrade the crystalline quality and surface morphology of the N-polar III-nitride films [10]. Therefore, some research groups have dedicated to improve the crystalline quality and surface morphology for N-polar GaN, AlN, and AlGaN films. By using a flow-rate-modulation technology with an interruption time of 4 s to grow the low temperature (LT)-GaN nucleation layer, the crystalline quality of the N-polar GaN was significantly improved [11]. The sapphire substrate with a misorientation angle of 2.0° was reported to be useful to grow the N-polar AlGaN film [12]. The growth of N-polar AlN layer with flow-modulation technology was also conducted, and the full width at half maximum (FWHM) values of 72 and 1,020

arcsec were attained for (002) and (100) XRD diffractions, respectively [13]. In addition, some efforts have been devoted to the formation of polarity-inversion domains for Al-polar AlN epi-layer. It was claimed that nitrogen atoms replace the topmost oxygen atoms in Al_2O_3 and form N–Al bilayer with the N layer upward to realize the polarity-inversion [14]. The control of the polarity was also achieved by using a patterned LT-AlN buffer layer [15]. In spite of these efforts mentioned above, there are no reports yet on the polarity-reversion from Al(Ga)-polar to N-polar for the AlGaN film grown on the AlN seeding layer by using the flow-modulation technology.

In this paper, the effect of the innovative flow-modulation technology developed in this work on the epitaxial growth and characteristics of the AlN seeding layer was studied extensively. The mechanism for the growth was revealed for the AlN seeding layer which turned out to play a critical role in the polarity reversion of the subsequently grown AlGaN film. Moreover, significant improvements in crystalline quality and surface morphology of the N-polar AlGaN film were achieved by carefully optimizing the V/III ratio for the epitaxial growth process.

2. Experimental

The N-polar AlGaN/AlN heterostructures were grown in a vertical low-pressure MOCVD system. 2-inch c (0001) plane sapphire substrates were used to grow all the samples in this study. Before the growth process, the sapphire substrates were heated up to 1,090 °C in hydrogen ambience for 2 min to clean the surface. Then nitridation process for the sapphire substrate was performed at 1,000 °C in ammonia (NH_3) ambience for 1 min, paving a thin layer of N atoms on the surface of the sapphire substrate. This is a key prerequisite for the subsequent growth of the N-polar III-nitrides. After the above pretreatment, the high temperature (HT)-AlN seeding layer was deposited by utilizing the newly-developed flow-modulation technology at 980°C with a reactor chamber pressure around 70 Torr. The flow rates of trimethylaluminum (TMA) and NH_3 for HT-AlN seeding layer were 9.3 $\mu\text{mol/min}$ and 180 mmol/min , respectively. TMA and NH_3 were injected into the reactor

chamber alternatively and periodically. Figure 1 shows the source supply chart of TMA and NH_3 for the growth of the HT-AlN seeding layer. For each source-supplying cycle, NH_3 was provided for 6 s without TMA flowing immediately after supplying TMA for 6 s without NH_3 flowing. The growth process for the N-polar HT-AlN seeding layer was completed in 150 cycles. The five N-polar AlGaIn film samples labeled as samples A, B, C, D, and E were grown with a fixed NH_3 flow rate of 45 mmol/min, but varied TMA flow rates and thus varied V/III ratios at 1,050 °C. In addition, an AlGaIn/AlN heterojunction sample named as sample F was grown with conventional continuous growth method.

The N-polar AlGaIn film can be corroded in 5 mol/l KOH solution at 100 °C in a few minutes whereas the metal-polar AlGaIn film can not be removed in such a solution. Therefore, the polarity of the grown AlGaIn film was verified in this study by etching with KOH solution. High-resolution X-ray diffraction (HR-XRD) measurement was performed to characterize the crystalline quality of N-polar AlGaIn film. The root mean square (RMS) value obtained from atomic force microscopy (AFM) was used to evaluate the surface roughness of the AlN seeding layer. The surface morphology of the N-polar AlGaIn film was investigated with optical microscope (OM) and scanning electron microscopy (SEM).

3. Results and discussion

To investigate the effect of the flow modulation on the surface morphology of the N-polar HT-AlN seeding layer, the AFM images of the HT-AlN seeding layers grown under continuous and flow-modulation modes were taken within a detection area of $5 \times 5 \mu\text{m}^2$ and shown in Figs. 2(a) and 2(b), respectively. The RMS values were measured from Fig. 2 to be 2.8 nm for the HT-AlN seeding layer grown under continuous mode, and 1.6 nm for the HT-AlN seeding layer grown under flow-modulation mode. With the introduction of the newly-developed flow-modulation technology, the remarkable improvement in surface morphology was ascribed to the enhancement in the migration capacity of Al atoms on the surface of the HT-AlN seeding layer. In fact, when only TMA flow and no NH_3 flow was

injected into the reaction chamber, which was equivalent to the reduction in the ammonia partial pressure simultaneously [16], there were just few N atoms remaining on the surface of AlN epitaxial layer. Thus the migration distance of Al atoms was long enough, surpassing the terrace width and forming the Al-N bonds actively as the NH_3 flow was delivered onto the surface of sapphire substrate once again [17]. The truncated formation of these AlN islands because of the diffusing first and reacting later sequence induced isotropic growth pattern for the HT-AlN seeding layer. There was denser nucleation domains observed on the surface of the HT-AlN seeding layer of the sample grown under flow-modulation mode than that grown under continuous mode. The triangular grains shown in Fig. 2(a) were probably originated from the anisotropy in migration rate for various kinds of atoms. When TMA and NH_3 flows were simultaneously applied onto the surface of sapphire substrate, the short migration length of Al atoms triggered three-dimensional growth of AlN film [18], resulting in the high density of streamlined grains as shown in Fig. 2(a). The surface morphology was also known to be influenced by the misorientation of the sapphire substrate or surface supersaturation [19]. The surface features including the N-region coverage are sensitive to the deviations from the normal situation of the reactor in the flow-modulation growth process [20]. It can be summarized that the surface roughness is strongly dependent on the polarity of the AlN epitaxial film and the exposed atmosphere as well.

The AlGaIn/AlN epi-layer samples C and F were etched with KOH solutions for 0, 6, and 12 min, respectively. Figures 3(a), 3(c), and 3(e) demonstrate the surface of the N-polar AlGaIn film sample C grown on the N-polar AlN seeding layer under the flow-modulated mode before (Fig. 3(a)), and after etching with KOH solution for 6 min (Fig. 3(c)) and 12 min (Fig. 3(e)), respectively. It is evident that the hexagonal cone pits usually observed on the surface of the N-polar AlGaIn film disappeared after 6 min corrosion in KOH solution as shown in Fig. 3(c). After etching with KOH solution for 12 min, the whole AlGaIn/AlN epitaxial film of sample F was nearly completely removed, and the sapphire substrate with a flat and smooth surface was

exposed to the air as shown in Fig. 3(e). The black dots on the image shown in Fig. 3(e) may be the residual Al-polar AlN grains that could exist at the initial growth stage of the N-polar HT-AlN seeding layer [21]. It was reported that the partial inversion of the polarity from N-polar to Al-polar in the growth process of the N-polar AlN film was a result of the decomposition of sapphire substrate in the presence of hydrogen [22]. In contrast, as shown in Figs. 3(b), 3(d), and 3(f), there was almost no any variation in the appearance of sample F grown under continuous mode before and after wet etching with KOH solution. This fact indicates that the AlGaN/AlN epi-layer sample F is Al(Ga)-polar. The remarkable variation in polarity could be attributed to the appropriate control of the NH_3 flow in the growth process of the AlN seeding layer with the innovative flow-modulation technology. The excessive supply of N atoms will promote the decomposition of the sapphire substrate, inducing the variation in the growth mode of the AlN seeding layer [23], thereby reverse the polarity of the AlGaN/AlN epi-layers.

The XRD $2\theta - \omega$ scanning curves for samples C and F are shown in Figs. 4(a) and 4(b), respectively. The XRD peaks of the AlGaN epi-layers from (0002) plane for both samples are located around 34.8 degree. As depicted in Fig. 4(a) in red dotted line, the XRD intensities of (0002) plane AlGaN- and AlN-epi-layer-related peaks for sample C grown under the newly-developed flow-modulation mode drop sharply after etching. However, as shown in Fig. 4(b), the XRD intensities of (0002) plane AlGaN- and AlN-epi-layer-related peaks do not change significantly even after etching for 12 min. This result demonstrates once again that the polarity reversion from Al(Ga)-polar to N-polar for the AlGaN/AlN epitaxial films can be realized by just growing the HT-AlN seeding layer with the flow-modulation technology developed in this study.

In order to investigate the impact of V/III ratio on the crystalline quality of the N-polar AlGaN film samples A-E, the HR-XRD rocking curves (XRCs) were measured. The FWHM values of the XRCs and the size of the hillock-like defects on the surfaces of the N-polar AlGaN films as a function of V/III ratio are plotted in Fig. 5 and summarized in Table 1. It was demonstrated clearly in Fig. 5 that with the

increase in V/III ratio, the FWHM value of the (0002) plane XRC for the N-polar AlGaIn film samples A-E varied in a fluctuating mode, and a FWHM value as small as 450 arcsec was achieved with sample B which was grown with a V/III ratio of 988.

Since the NH₃ flow rate was set to be a constant of 45 mmol/min, the increase in the V/III ratio meant a decrease in both TMA and TMG flow rates. It is well known that both the flow rates and the diffusion rates of TMA and TMG have remarkable influence on the atom deposition [24]. The molarity of an ideal precursor gas is only associated with its partial pressure in a MOCVD reactor chamber with a fixed volume at certain growth temperature, whereas the partial pressure of the gas-phase chemistry will affect the overall epitaxial growth process. There are two competing routes for the gas-phase group-III nitride chemistry, namely the adduct formation pathway and the thermal decomposition pathway [25]. It was reported that the methyl radicals generated in the decomposition process of TMA needed more energy than TMG [26]. Therefore, it is more difficult for TMA to be decomposed than TMG. In this study, the thermal decomposition reaction for TMA/TMG that occurred near the substrate surface at high temperature can be expressed as [27, 28]

(1)

(2)

(3)

(4) Here $X = Al \text{ or } Ga$. The TMA flow rates used to grow samples A and B were relatively large as shown in Table 1 so that the thermal decomposition rather than the adduct formation dominated the reaction near the surface of the N-polar AlGaIn epitaxial film. Moreover, the chemical reaction equilibrium moved forward with the increase in the TMA/(TMA+TMG) molar ratio from 0.29 for sample A to 0.32 for sample B, further promoting the thermal decomposition reaction. The formation of the

N-polar AlGa_N film can be expressed as

where α is the Al composition. The diffusion rates of both Al and Ga atoms slowed down because of the mass atomic aggregation, which was beneficial to the formation of Al(Ga)-N bonds and thus to the improvement in crystalline quality of the N-polar AlGa_N film. This is why the crystalline quality of sample B was better than sample A. However, the adduct formation would predominate when the TMA flow rates used to grow samples C, D, and E were small. The adduct formation occurred in the mixture of TMG, TMA, and NH₃ was generally considered to be a “parasitic” reaction and could be given by

where β is the Al composition. With a fixed TMA/(TMA+TMG) molar ratio of 0.32 and a decrease in TMA flow rate from 14.6 $\mu\text{mol/min}$ for sample B to 11.7 $\mu\text{mol/min}$ for sample C, the adduct-related nanometer-scale particles were introduced during the adduct formation, leading to an abrupt deterioration in crystalline quality of the N-polar AlGa_N film. On the other hand, the TMA/(TMA+TMG) molar ratio decreased from 0.32 for sample C to 0.31 for sample D. As a result, the chemical reaction equilibrium expressed by Eq. (6) moved in opposite direction, slowing down the growth rate and thus improving the crystalline quality of the N-polar AlGa_N film. The TMA/(TMA+TMG) molar ratio increased from 0.31 for sample D to 0.35 for sample E, which could accelerate the growth rate and degrade the crystalline quality of the N-polar AlGa_N film again. In fact, since the adduct formation is unavoidable during the MOCVD growth process for the group-III nitrides, the crystalline quality will be degraded by the adducts due to the impairment of uniformity and the extra consumption of the feeding stream of the precursors [29]. When the thermal decomposition pathway was dominant in the deposition process, the smallest FWHM value and the best crystalline quality were obtained with sample B which was grown at a V/III ratio of 988. Therefore, the physical mechanisms behind the observed fluctuating behavior of the FWHM values of the N-polar AlGa_N epi-layer samples

shown in Fig. 5 have been revealed in terms of the V/III ratio and TMA/(TMA+TMG) molar ratios. Our results also imply that there should be a certain balanced or optimized MOCVD growth condition for achieving the N-polar AlGa_N films with high crystalline quality.

The maximum diagonal length of the hillock-like defect region obtained from the SEM measurement is defined as the defect size, and plotted as a function of V/III ratio in Fig. 5. The defect size decreased monotonously with the increase in V/III ratio although the crystalline quality was not improved correspondingly as mentioned above. Figure 6 shows the top-view SEM images of the N-polar AlGa_N films grown with varied V/III ratios. It is evident that the three-dimensional growth mode-induced hexagonal cone-like defects were usually generated when the N-polar AlGa_N film was grown at relatively low V/III ratio as shown in Figs. 6(a) and 6(b). This phenomenon can be attributed to the oversupply of group-III precursors, leading to the excess of Al atoms in the MOCVD reactor chamber during the thermal decomposition reaction. On the other hand, the increase in the number of nucleated grains with the increase in the V/III ratio is significant as demonstrated clearly in Figs. 6(c), 6(d) and 6(e). These small white dot-like crystal nuclei appearing on the terraces could trigger the generation of hillock-like defects during the adduct formation reaction. Furthermore, they promote the “islands” merging in a two-dimensional (2D) pattern, causing the transition of the growth mode for N-polar AlGa_N films from three-dimensional (3D) to 2D.

The first-order derivatives (FOD) of the relative light transmission (RLT) determined from the UV-visible absorption spectra (not shown here) for the N-polar AlGa_N film samples A-E are demonstrated in Fig. 7. Based on the peak values obtained from Fig. 7, the Al compositions for the N-polar AlGa_N films was calculated. An Al composition as high as 21% was achieved for sample E. With the increase in the V/III ratio, the outstanding blue shift of the FOD peaks denoted in Fig. 7 with black dotted line was due to the increase in incorporation efficiency of the Al atoms into the lattice.

4. Conclusions

The MOCVD growth, the polarity reversion, surface morphology, and the crystalline quality of the N-polar AlGa_N films deposited on the specific AlN seeding layers with an innovative flow-modulation technology were studied extensively. The polarity reversion from Al(Ga)-polar to N-polar for the AlGa_N epi-layer could be realized by growing the AlN seeding layer with the newly-developed flow-modulation technology. Moreover, a FWHM value of (0002) XRC as small as 450 arcsec was achieved for the N-polar AlGa_N film grown with a V/III ratio of 988. Meanwhile, the size of the hexagonal cone-like defects on the surface of the N-polar AlGa_N film was found to decrease monotonously with the increase in V/III ratio. Furthermore, the observed fluctuating behavior of the FWHM values of the N-polar AlGa_N epi-layer samples as a function of the V/III ratio was explained in terms of the molar ratio TMA/(TMA+TMG). This ratio determines the migration of Al and Ga atoms in gas-phase chemistry routes including the adduct formation pathway and the thermal decomposition pathway.

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Fig. 1. The source supply chart of TMA and NH_3 for the growth of the HT-AlN seeding layer with the newly-developed flow-modulation technology.

Fig. 2. The AFM images taken within a detection area of $5 \times 5 \mu\text{m}^2$ on the surface of the HT-AlN seeding layers grown under continuous (a) and flow-modulation (b) modes.

Fig. 3. The OM images for the surfaces of the AlGaIn/AlN epi-layer samples C and F deposited on the HT-AlN seeding layers grown in flow-modulation mode and continuous mode before (a, b) and after etching with KOH solution for 6 min (c, d) and 12 min (e, f), respectively.

Fig. 4. The (0002) $2\theta - \omega$ XRD scanning curves for the AlGaIn/AlN epi-layer samples C (a) and F (b) measured before (black solid lines) and after (red dotted lines) etching with KOH solution.

Fig. 5. The FWHM values of the (0002) plane XRCs and the sizes of the hillock-like defects on the surfaces of the N-polar AlGaIn film samples A-E as a function of the V/III ratio.

Fig. 6. The top-view SEM images of the N-polar AlGaIn film samples A-E grown with various V/III ratios.

Fig. 7. The first-order derivative (FOD) of the relative light transmission (RLT) in the UV-visible absorption spectra for the N-polar AlGaIn film samples A-E.

Sample	TMA flow rate ($\mu\text{mol/min}$)	TMA/(TMA +TMG) molar ratio	V/III ratio	Al composition (percentage)	(0002) FWHM (arcsec)	Defect size (nm)
A	26.2	0.29	499	8.98	594	6.43
B	14.6	0.32	988	14.45	450	5.35
C	11.7	0.32	1236	17.20	691	2.73
D	9.3	0.31	1502	18.30	527	2.55
E	8.7	0.35	1782	21.06	677	1.61

Table1. Summary of the TMA flow rates, TMA/(TMA+TMG) molar ratios, the V/III ratios, the Al compositions, the (0002) plane FWHM values of the XRCs as well as the sizes of surface hillock-like defects for the N-polar AlGa_N/AlN epi-layer samples A-E.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Sample	A	B	C	D	E
V/III ratio	499	988	1236	1502	1782

Fig. 6

Fig. 7

Conflicts of Interest

Dear Editor,

We declare that we have no financial and personal relationships with other people or organizations that can inappropriately influence our work, there is no professional or other personal interest of any nature or kind in any product, service and/or company that could be construed as influencing the position presented in, or the review of, the manuscript entitled, “Achievement of polarity reversion from Al(Ga)-polar to N-polar for AlGa_N film on AlN seeding layer grown by a novel flow-modulation technology”.

Title: Achievement of polarity reversion from Al(Ga)-polar to N-polar for AlGa_N film on AlN seeding layer grown by a novel flow-modulation technology

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Thank you very much for your time and effort on our matter. We are looking forward to hearing from you soon.

Sincerely yours,

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Figures

Figure 1

The source supply chart of TMA and NH_3 for the growth of the HT-AlN seeding layer with the newly-developed flow-modulation technology.

Figure 2

The AFM images taken within a detection area of $5 \times 5 \mu\text{m}^2$ on the surface of the HT-AlN seeding layers grown under continuous (a) and flow-modulation (b) modes.

Figure 3

The OM images for the surfaces of the AlGaIn/AlN epi-layer samples C and F deposited on the HT-AlN seeding layers grown in flow-modulation mode and continuous mode before (a, b) and after etching with KOH solution for 6 min (c, d) and 12 min (e, f), respectively.

Figure 4

The (0002) 2θ - ω XRD scanning curves for the AlGaN/AlN epi-layer samples C (a) and F (b) measured before (black solid lines) and after (red dotted lines) etching with KOH solution.

Figure 5

The FWHM values of the (0002) plane XRCs and the sizes of the hillock-like defects on the surfaces of the N-polar AlGaIn film samples A-E as a function of the V/III ratio.

Sample	A	B	C	D	E
V/III ratio	499	988	1236	1502	1782
2μm	2μm	2μm	2μm	2μm

Figure 6

The top-view SEM images of the N-polar AlGaIn film samples A-E grown with various V/III ratios.

Figure 7

The first-order derivative (FOD) of the relative light transmission (RLT) in the UV-visible absorption spectra for the N-polar AlGaIn film samples A-E.