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Effect of indium accumulation on the characteristics of a-plane InN epi-films under different growth conditions

Yun-Yo Lo [a], Man-Fang Huang [a,*], Yu-Chia Chiang [a], Jenn-Chyuan Fan [b]

[a] Institute of Photonics, National Changhua University of Education, Changhua, Taiwan, ROC
[b] Department of Electronic Engineering, Nan Kai University of Technology, Nantou, Taiwan, ROC

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ABSTRACT

This study investigated the influence of indium accumulation happened on the surface of a-plane InN grown under different growth conditions. Three different growth rates with N/In ratio from stoichiometric to N-rich were used to grow a-plane InN epifilms on GaN-buffered r-plane sapphires by plasma-assisted molecular beam epitaxy. When a-plane InN was grown above 500 °C with a high growth rate, abnormally high *in-situ* reflectivity was found during a-plane InN growth, which was resulted from indium accumulation on surface owing to In-N bonding difficulty on certain crystal faces of a-plane InN surface. Even using excess N-flux, indium accumulation could still be found in initial growth and formed 3-dimension-like patterns on a-plane InN surface which resulted in rough surface morphology. By reducing growth rate, surface roughness was improved because indium atoms could have more time to migrate to suitable position. Nonetheless, basal stacking fault density and crystal anisotropic property were not affected by growth rate.

1. Introduction

III-nitrides with fine quality, commonly grown on c-plane sapphire or other expensive substrates, have been obtained [1–7]. Among III-nitrides, InN has the advantages of small direct bandgap of 0.7 eV, high electron mobility and saturation velocity [8,9]. However, high quality InN epifilm is difficult to grow due to low dissociation temperature and lack of appropriate substrates. Moreover, InN and others III-nitride are wurtzite crystal structures having spontaneous polarization along the polar c-axis in epi-layers and piezoelectric polarization induced from hetero-interface lattice mismatch. The total polarization leads to quantum confined Stark effect (QCSE) in active layer [10] and affects the performance of optoelectronic devices. In addition, electron accumulation was present on the polar nitride surface but absent on nonpolar nitride surface. To avoid QCSE and electron accumulation, the growth of nonpolar III-nitrides, such as (1–100) m-plane and (11–20) a-plane, has been investigated [11–19].

Compared with m-plane III-nitride, a-plane III-nitride can be grown on r-plane sapphire that is cheap and has high quality. However, the quality of a-plane III-nitride epi-film is usually worse than that of c-plane III-nitride epi-film, which is caused by anisotropic structure on growth surface and un-mature growth condition. The anisotropic crystalline property was found from the differences in full-width at half maximum (FWHM) of on-axis X-ray rocking curve (XRC) measured at different azimuth angles [17–20]. Previous a-plane GaN studies demonstrated that the FWHM of XRC at different azimuth angles could be changed when a-plane GaN epi-films were grown using different growth conditions and buffer layers [19–21]. Similar study for a-plane InN was also reported [22], which showed that the degree of the structural anisotropy could be minimized by combining high nitridation temperature and high growth temperature.

In addition, several groups have dedicated to improve crystal quality and surface morphology for either polar or nonpolar InN by employing different nitridation procedures [23], various buffer layers, growth temperatures [11–16,24] and V/III ratios [24]. For growing a-plane InN, a-plane GaN layer grown on r-plane sapphire is usually used as a buffer layer since epi-growth of a-plane GaN on r-plane sapphire has been intensively studied. Moreover, a-plane InN grown on a-plane GaN template was demonstrated [14,25]. Crystalline quality and surface roughness of a-plane InN were improved because a-plane GaN template grown by metal-organic vapor phase epitaxy had a better quality than molecular beam epitaxy (MBE)-grown a-GaN buffer layer. In addition, if V/III ratio was slightly metal-rich, crystalline quality and surface morphology of a-plane InN could be further improved because a few monolayers of indium was covered on the surface of a-plane InN epifilm [14]. Moreover, with the use of low-temperature InN buffer layer, surface morphology could be further improved even if growth temperature was high [13]. Nonetheless, crystalline quality of a-plane InN was still inferior to that of a-plane GaN. Except growth temperature, V/III ratio and buffer layer, how growth rate influences a-plane InN quality is still unknown. In this paper, the characteristics of a-plane InN grown

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* Corresponding author.
E-mail address: mfhuang@cc.ncue.edu.tw (M.-F. Huang).

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on GaN-buffered r-plane sapphire with various growth rates and V/III ratios are reported. The influence of indium accumulation happened on a-plane InN surface under different growth conditions will be discussed.

2. Experiment

In this study, a-plane InN was grown by plasma-assisted MBE on r-plane sapphire. Before growing a-plane InN, r-plane sapphire was thermal-cleaned at high temperature followed by high-temperature nitridation. After nitridation, a 20-nm N-rich GaN buffer followed by a 30-nm stoichiometry GaN buffer was grown on nitridized r-plane sapphire. Then a-plane InN epifilm was grown at 525 °C for 1 h on GaN-buffered r-plane sapphire with various N/In ratios and growth rates (GRs). Growth temperature we chose in this study was close to the upper limit of growth temperature for N-face c-plane InN. Five samples were grown in this study. Detailed growth conditions including indium beam equilibrium pressure (BEP), N_2 flow and radio frequency (RF) power were tabulated in Table 1. N/In ratios (i.e. V/III ratios) in N-rich condition were estimated from reduction percentage of In flux compared with the In flux used in stoichiometric condition. Among these five samples, samples A and B had the same low GR of 295 nm/h with the same In flux, and their N/In flux ratios were 1 and 1.2. Samples B, C, and E had the same N flux (1 sccm N_2 and RF-power of 240 W). Their GRs and N/In ratios were controlled by changing In flux. Samples E and D had the same high GR of 350 nm/h and similar N/In ratios of 1 and 1.2 but had a higher In flux compared with other samples. During epitaxial growth, GR and surface roughness were monitored from 470-nm *in-situ* reflectance interferometry of SVT4000 pyrometer. After epi-growth, surface morphology was observed by atomic force microscope (AFM) of NT-MDT Solver P47, operated in the tapping mode, and Hitachi 4700 scanning electron microscopy (SEM) with an operating voltage of 15 KV. High resolution X-ray diffraction (XRD) ω-2θ scan and XRC were taken using Siemens D 5000 X-ray diffractometer to characterize crystalline quality.

3. Results and discussions

The crystalline quality of a-plane InN samples was first examined by XRD. Fig. 1 shows XRD ω-2θ scan results of samples A, B and E. The XRD results of samples C and D were similar and not shown here. As indicated in Fig. 1, only a-plane GaN (57.77°), a-plane InN (51.62°) and r-plane sapphire (25.57° and 52.54°) diffraction peaks were obtained. Indium droplet, (101)-face InN (32.9°) or other crystal faces were not found. This indicated that all samples grown on GaN-buffered r-plane sapphire were pure a-plane single-crystal InN films. Moreover, FWHM values of XRC along c-orientation and m-orientation were around 0.5° and 1°, respectively, for all samples. Anisotropic crystalline quality can be also found from Fig. 2 which shows plots of XRC FWHM of (11–20) plane at different azimuth angles for samples A, B and E. Samples A and B had the same GR but different N/In ratios. Samples A and E had the same N/In ratios but different GR. Because these three samples had almost the same patterns, it indicated that anisotropic crystalline quality of a-plane InN was not affected by N/In ratios and growth rate.

Next the surface morphologies of all samples were examined by SEM and AFM. The surface morphologies of a-plane InN with different N/In ratios are first compared. Fig. 3 shows SEM surface images of samples A and B grown with the same low GR but different N/In ratios. As shown in Fig. 3(a), sample A grown under stoichiometric growth condition (i.e. N/In ratio = 1) has rhombus holes on its epifilm surface. However, as shown in Fig. 3(b), sample B grown under N-rich condition (with In/N ratio of 1.2) has stripe-like patterns on surface. Either stripes or rhombus holes shown on the surface of a-plane InN are mainly caused by anisotropic crystalline properties of a-plane InN because the GR of InN is increased along [0001] direction but suppresses along the [10–33] direction. Similar phenomena were found in previous InN nanocolumn [26] and a-plane GaN [27] studies. Although N-rich condition leads to more stripe-like morphology on the surface of a-plane InN compared with stoichiometric growth condition. However, anisotropy of a-plane InN, indicated from XRD results, remains the same and is not affected by V/III ratios. More details will be discussed later.

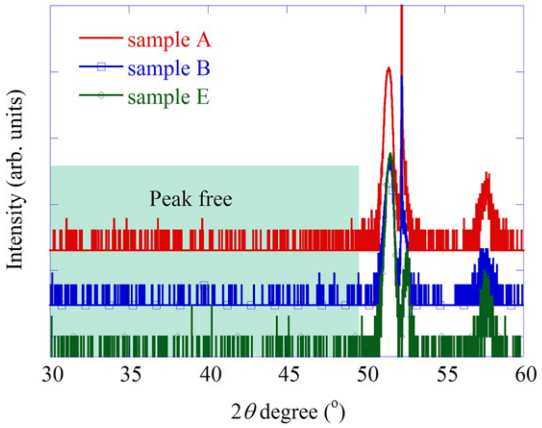

Fig. 1. Symmetrical XRD 2θ-scan profiles for samples A, B and E. The incident plane is parallel to [0001] axis.

Table 1
Summary of growth conditions and roughness RMS values for all a-plane InN samples.

Sample	GR (nm/h)	Indium BEP (10^{-4} Pa)	N_2 flow (sccm)	RF power (W)	N/In ratio	RMS (nm)
A	295	1.25	0.8	240	1	2.6
B	295	1.25	1	240	1.2	5.97
C	310	1.33	1	240	1.13	7.65
D	350	1.50	1	300	~1.2	8.73
E	350	1.50	1	240	1	10.05

Moreover, the surface morphologies of a-plane InN with different GRs are then compared from AFM images. Fig. 4 shows 3 × 3 μm^2 AFM images of samples B, C, and E. These three samples were grown with the same N_2 flow but different In flux, i.e. different N/In ratios and GRs. As indicated in Fig. 4(a), the surface of sample B grown with low In flux and GR (i.e. high N/In ratio), has stripe-like patterns, which is similar to the SEM image shown in Fig. 3. The AFM image of sample A with same GR also has similar patterns (the small picture in Fig. 4(a)), although those stripes are obscure. Moreover, as shown in Fig. 4(b), some regular stripes are also observed on the surface of sample C grown with increased In flux and GR (i.e. slightly N-rich condition

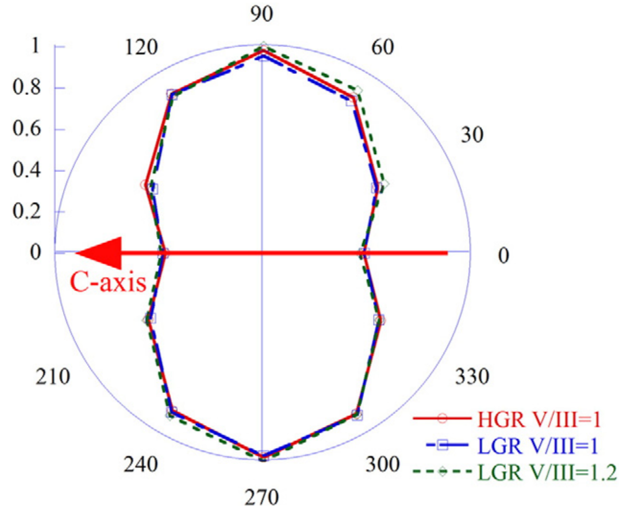

Fig. 2. FWHMs of (11–20) XRC with different azimuth angles.