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MOVPE growth of InGaN on Si(111) substrates with an intermediate range of In content

Ashraful G. Bhuiyan[1], A. Mihara[1], T. Esaki[1], K. Sugita[*, 1], A. Hashimoto[1], A. Yamamoto[, 1], N. Watanabe[2], H. Yokoyama[2], and N. Shigekawa[2]**

[1] Graduate School of Engineering, University of Fukui, 3-9-1 Bunkyo, Fukui 910-8507, Japan
[2] NTT Photonics Laboratories, 3-1 Morinosato Wakamiya, Kanagawa 243-0198, Japan

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* Corresponding author: e-mail k_sugita@u-fukui.ac.jp, Phone: +81-776-27-8566, Fax: +88-776-27-8749
** e-mail ayamamot@u-fukui.ac.jp, Phone: +81-776-27-8566, Fax: +88-776-27-8749

This paper reports on the MOVPE growth of InN and $In_xGa_{1-x}N$ with x up to ~0.4 on AlN/Si (111) substrates. Good quality InN films are grown after a pretreatment of the substrate surface at 1000 °C in NH_3. While pretreatment of the substrate surface in N_2 results the In droplet formation. Single-crystalline $In_xGa_{1-x}N$ films with x up to ~0.4 are successfully grown on Si (111) substrates without phase separation and metallic In incorporation by changing the growth temperature and TMI/(TMI+TEG) molar ratio. Comparative study between the AlN/Si (111) and GaN/α-Al_2O_3 (0001) substrates shows that the quality of InN, GaN and InGaN with low In contents depends on the quality of the underlying layer. While the quality of InGaN at the intermediate In content range hardly depends on of the underlying layer/substrate. These works will allow us towards the fabrication of the future InGaN-Si tandem cell using MOVPE.

1 Introduction The $In_xGa_{1-x}N$ compound semiconductors have been predicted as excellent candidates for full-solar spectrum photovoltaic (PV) applications. This was based on their suitability to the engineering of band gap energies from 0.64 to 3.4 eV with characteristics of direct band gap, high absorption coefficients, and excellent irradiance resistance [1]. Recently, $In_{0.45}Ga_{0.55}N$ is proposed as an appealing candidate to be paired with Si in tandem solar cells that may have power conversion efficiency higher than 31% [2]. A low resistance ohmic junction is predicted for the n-$In_{0.45}Ga_{0.55}N$/p-Si junction and, therefore, no tunnel junction is necessary to connect the top and bottom cells [3]. Therefore, InGaN materials have plenty of opportunity for higher efficiencies where most competing technologies are very near to their maximum practical efficiencies.

Most of the recent InGaN-based solar cells have been grown on sapphire or GaN templates. However, Si is a better choice either as substrate material or as a bottom cell for the InGaN-based solar cells. Because, large size Si is widely available with very low cost. Since Si is semiconductor, formation of vertical contact can easily be made without mesa structure as for sapphire substrates. Good quality GaN and InN layers have been grown on Si substrates [4]. However, the growth of $In_xGa_{1-x}N$ with $x>0.1$ on Si remains a challenging task [5]. Recently, MBE growth of $In_xGa_{1-x}N$ with x up to ~0.3 has been reported [6]. There have been no reports on the MOVPE growth of InGaN on Si especially with an intermediate range of In content. In this work, we have successfully grown single-crystalline InN and $In_xGa_{1-x}N$ with x up to ~0.4 on AlN/Si (111) substrate by MOVPE.

2 Experimental Using a MOVPE system with a horizontal reactor, InN and InGaN films are grown on AlN/Si (111) and GaN/sapphire substrates. Prior to the growth, the substrate surfaces are annealed in N_2 or NH_3 atmosphere at a different temperature and time. As precursors, triethylgallium (TEG), trimethylindium (TMI), and NH_3 are used. The InN epilayer is grown at 600 °C in a pressure 760 Torr. For the InGaN epilayer growth, the pressure is fixed at 150

Torr and the temperature is changed from 600 to 850 °C. To control the In composition in InGaN, both TMI/(TMI+TEG) molar ratio and growth temperature are varied and optimized. To estimate In composition and evaluate crystalline quality, X-ray diffraction of $2\theta/\omega$ and ω methods are employed.

3 Results and discussions Technologies for the growth GaN on Si substrates have been matured while the InN and In-rich InGaN have not yet been established. Before growing InGaN we first attempted to grow InN on AlN/Si (111) substrates. Figure 1 shows the XRD 2θ/ω profiles of InN films grown on AlN/Si (111) substrate with different substrate pretreatments at 1000 °C. As shown in Fig. 1, substrate pretreatments in NH_3 made possible to grow InN without In droplets formation while substrate pretreatments in N_2 resulted in formation of In droplets in the grown film. It seems to be related with the change of AlN surface polarity from Al- to N-polar as a result of annealing in NH_3 atmosphere, which resulted in the growth of N-polar InN film and suppressed the In incorporation in the grown film. Substrate pretreatment in NH_3 results better quality than in N_2. The quality of the InN films on Al/Si (111) substrates is found to be similar with the InN films on GaN/α-Al_2O_3 (0001). Long time pretreatment is found to be deteriorated the substrate surface and then epilayer quality.

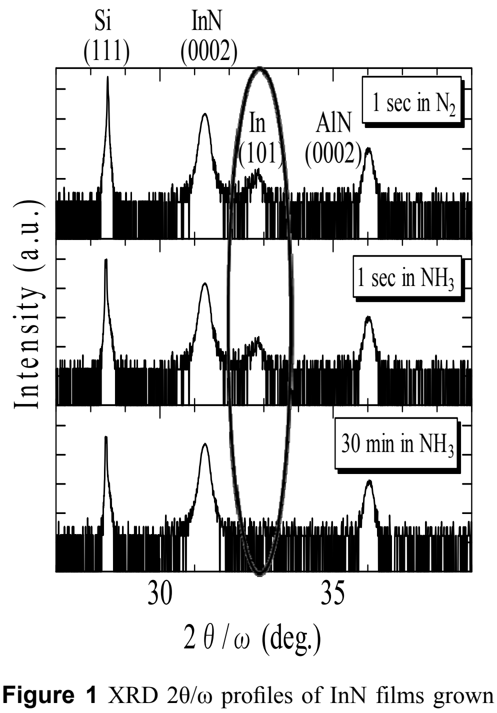

Figure 1 XRD 2θ/ω profiles of InN films grown on AlN/Si(111) substrate with different substrate pretreatments at 1000 °C.

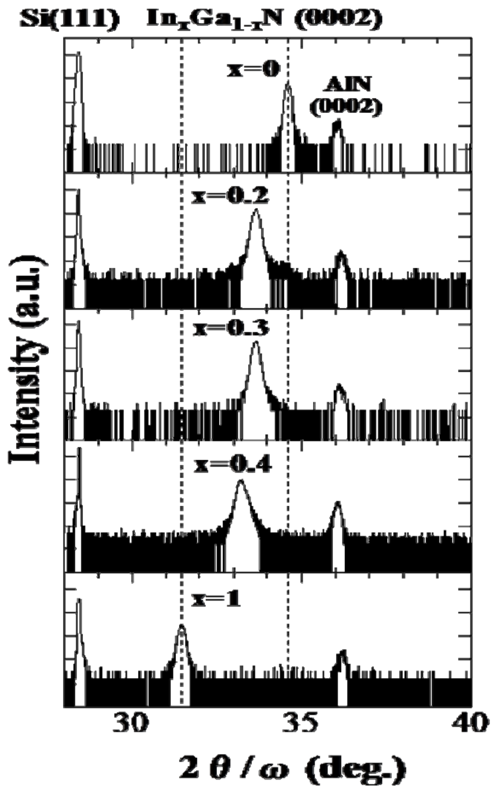

Figure 2 XRD 2θ/ω profiles of $In_xGa_{1-x}N$ films grown on AlN/Si(111) substrate with different In contents.

Figure 2 shows the XRD 2θ/ω profiles of $In_xGa_{1-x}N$ films grown on AlN/Si (111) substrate with different In contents. The In content is changed by varying the growth temperature and the TMI/(TMI+TEG) molar ratio as shown in Fig. 3. Single crystalline $In_xGa_{1-x}N$ films with x up to ~0.4 on AlN/Si (111) substrate are successfully grown without phase separation and In droplet formation as shown in Fig. 2. In order to evaluate the crystalline quality of the InGaN films grown on AlN/Si (111), XRC-FWHM (tilt fluctuation) is measured and shown in the Fig. 4. The XRC-FWHM for the InGaN films grown on GaN/α-Al_2O_3 (0001) is also shown for comparison. The quality of the grown InGaN films is found to be better on GaN/ α-Al_2O_3 than on AlN/Si. This is because the XRC-FWHM of the underlying GaN layer is 4.9 arcmin while for the AlN layer it is 28.8 arcmin. The quality of the grown InGaN films is deteriorated on both the substrates with increasing the In content and the difference in the crystalline quality of the InGaN films grown on both substrates becomes very small at an In content around 0.4, as shown in Fig. 4.

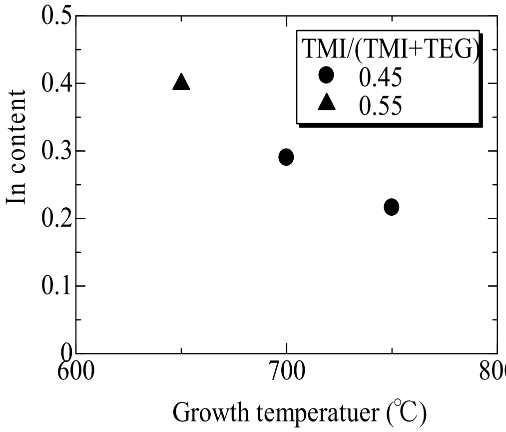

Figure 3 In content in InGaN films as a function of growth temperature as a parameter of TMI/(TMI+TEG) molar ratio.

These results indicate that the quality of InN, GaN and InGaN with low In contents depends on the quality of the underlying layer while it hardly depends on the quality of the underlying layer/substrate at the intermediate In content range for InGaN. This due to the increase of solid phase miscibility gap by increasing the In content as a result of large difference in interatomic spacing between InN and GaN. The stresses of the grown InGaN films on AlN/Si (111) are evaluated from the lattice parameters for an In content of 0.2 and 0.4 and shown in Table 1. At the In content 0.2, tensile and compressive stresses are found on Si and α-Al_2O_3, respectively, as expected. However, the stress is reduced at the In content 0.4, which is under investigation.

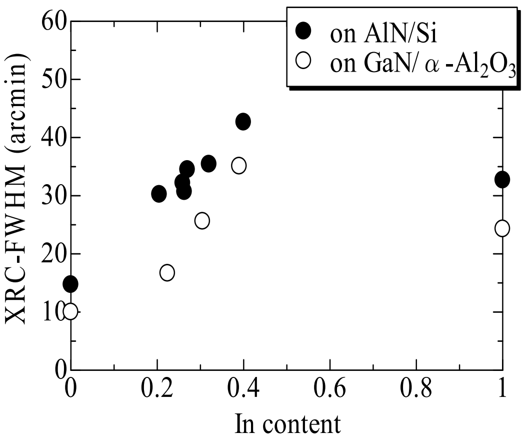

Figure 4 XRC-FWHM in InGaN films grown on AlN/Si (111) and GaN/α-Al_2O_3 (0001) substrates as a function of In content.

Table 1 Evaluated stress from InGaN films grown on AlN/Si (111) with an In content of 0.2 and 0.4.

Grown Film	Substrate	Stress evaluated from lattice parameters (GPa)
$In_{0.2}Ga_{0.8}N$ (Growth Temp. 700℃)	Si (111)	Tensile 0.6
	α-Al_2O_3(0001)	Compressive 0.4
$In_{0.4}Ga_{0.6}N$ (Growth Temp. 650℃)	Si (111)	Tensile 0.2
	α-Al_2O_3(0001)	Tensile 0.07

4 Conclusions MOVPE growth of InN and $In_xGa_{1-x}N$ with x up to ~0.4 on AlN/Si (111) substrates has been studied. Good quality InN films are grown after a pretreatment of the substrate surface at 1000 °C in NH_3. While pretreatment of the substrate surface in N_2 results the In droplet formation. Single-crystalline $In_xGa_{1-x}N$ films with x up to ~0.4 are successfully grown on Si (111) substrates without phase separation and metallic In incorporation by changing the growth temperature and TMI/(TMI+TEG) molar ratio. The quality of the grown InN, InGaN with low In contents and GaN is found better on the GaN/α-Al_2O_3 than on the AlN/Si, because of the good quality underlying layer of GaN when compared to the AlN. The quality of the grown InGaN films is deteriorated on both the substrates with increasing the In content and does not depend much on the underlying/substrate at the intermediate range. At the In content 0.2, tensile and compressive stresses are found on Si and α-Al_2O_3, respectively, as expected. However, the stress is reduced at the In content 0.4.

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