The Role of Ga to Improve AlN-Nucleation Layer for $Al_{0.1}Ga_{0.9}N$/Si(111)

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***Abstract*—**Group-III nitride material as particularly $Al_xGa_{1-x}N$ is one of promising optoelectronic materials to require for short-wavelength devices. To achieve the high-quality $Al_xGa_{1-x}N$ films for a high performance of such devices, AlN-nucleation layers are the important factor. To improve the AlN-nucleation layers with a variation of Ga-addition, XRD measurements were conducted to analyze the crystalline quality of the subsequent $Al_{0.1}Ga_{0.9}N$ with the minimum ω-FWHMs of (0002) and (10-10) reflections of 425 arcsec and 750 arcsec, respectively. SEM and AFM measurements were performed to observe the surface morphology and TEM measurements to identify the microstructures and orientations. Results showed that the optimized Ga-atoms in the Al(Ga)N-nucleation layers improved the surface diffusion to form more-uniform crystallites in structure and size, better alignment of each crystallite, and better homogeneity of island distribution. This, hence, improves the orientation of epilayers on the Si-surface and finally improves the crystalline quality and reduces the residual strain of subsequent $Al_{0.1}Ga_{0.9}N$ layers.

***Keywords*—**AlGaN, UV-LEDs, seed layers, AFM, TEM

I. INTRODUCTION

RECENTLY, $Al_xGa_{1-x}N$ is required as an optoelectronic material for short-wavelength light emission and absorption in many applications, e.g., purification, bio-chemical detectors and lighting [1,2] due to a direct bandgap energy ranging from 3.4 to 6.2 eV. To obtain a good material quality, the first prerequisite for such devices is by the deposition on a graded buffer layer and a good-quality nucleation layer (NL) etc [3,4]. In the case of AlGaN/GaN high electron mobility transistors (HEMTs), GaN layers grown on AlN NL showed a relationship between AlN NL strain and HEMT 2DEG properties with a better Hall sheet resistance by optimum layers [5]. To optimize the GaN layers, some groups have investigated the role of trimethylgallium flow during GaN nucleation-layer deposition and found that the optimal TMGa flow during the nucleation-layer growth leads to GaN films with superior structural and electronic properties [6]. Recently, there is a report on the optimum Al pre-deposition time to improve the crystal quality of AlN buffer layer and smoother surface with a reducing RMS roughness and rougher surface morphology of GaN layer in areas of the overlong Al-deposition time [7]. Here we have investigated the impact of Ga-atoms to improve the AlN-nucleation layers in order to improve the crystalline quality of $Al_{0.1}Ga_{0.9}N$ on AlN/Si(111) substrates.

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II. EXPERIMENTAL

Grown by an AIXTRON 200/4 RF-S MOVPE machine, the sample structure of the first series is ~300-nm $Al_{0.1}Ga_{0.9}N$ on ~140-nm AlN-buffer layer / ~10-nm Al(Ga)N NL with varied Ga-flow rate. The second is prepared with only one layer of Al(Ga)N-nucleation layers with a thickness variation and a variation of Ga-flow rate. In addition, the last series is a very thin layer of Al(Ga)N pre-deposition layer with a variation of Ga-flow rate. Trimethyl-gallium (TMGa), Trimethyl-aluminium (TMAl), and ammonia (NH_3) are used as precursors of Ga, Al and N, respectively. The growth parameters are maintained at 100 mbar and 1145°C, 1200°C. With an increasing triethyl-gallium (TEGa) flow rate in the Al(Ga)N- nucleation layer from 0 to 19.04 μmol/min, Seifert XRD3003HR and Seifert URD6 GID diffractometers were used to evaluate the crystalline quality, scanning electron microscopy (SEM) measurements and atomic force microscopy (AFM) measurements to observed surface morphology by Hitachi S4800 FE-SEM and Asylum Research MFP-3D-Bio AFM, respectively. Finally, JEOL JEM-2010 transmission electron microscopy (TEM) was performed to investigate cross-sectional microstructures and identify the orientation.

III. RESULTS

In Fig.1, XRD measurements show that ω-FWHMs of the $Al_{0.1}Ga_{0.9}N$ (0002) and (10-10) reflections decrease to the minimum of 425 arcsec and 750 arcsec, respectively at TEGa flow rate of 9.52 μmol/min and then increasing with an increasing TEGa flow rate in the Al(Ga)N-nucleation layer from 0 to 14.28 μmol/min. With decreasing ω-FWHM values, it means to the reducing tilt- and twist- misorientations of epilayer films. The optimized Ga flow rate into the AlN-

nucleation layer slightly improves the crystalline quality of subsequent $Al_{0.1}Ga_{0.9}N$ layers. Additionally, Ga in the AlN layers enables to decrease residual (in-plane) strain in the $Al_{0.1}Ga_{0.9}N$ layers and one reason is probably a lower lattice-mismatch of AlGaN than that of AlN.

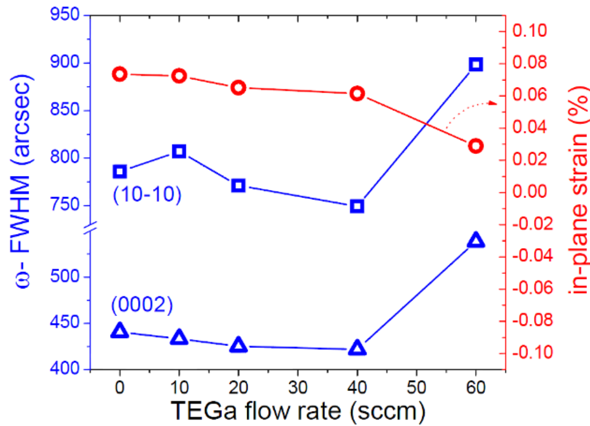

Fig. 1 The quality of subsequent $Al_{0.1}Ga_{0.9}N$ layers on AlN-nucleation layers with a variation of Ga flow rates

In Fig. 2, SEM measurements exhibit the surface morphology of the Al(Ga)N layers with an increasing growth time as shown in figures from above to below. Compared with the surface morphology of the AlN layers without Ga-addition (left-hand side), the Ga-added AlN layers (right-hand side) represent smaller and more-uniform crystallites, better homogeneity of AlN-island distribution, faster lateral coalescence and faster growth rate.

Moreover the growth rate and distribution of hexagonal-structure crystallites of the AlGaN pre-deposition layers with the Ga addition was improved better than that of the AlN pre-deposition layers as shown in Fig 2(a) and 2(d).

This implies to Ga atoms promoting to form the crystallites in the hexagonal structure. Subsequently, the AlGaN layers grown on the nucleation layers with the higher crystalline quality would be the better crystalline quality such as lower tilt- and twist- misorientations.

With a comparison to the SEM measurements, AFM measurements analogously reveal the surface morphology of the pre-deposition AlN layer with Ga-addition as smaller crystallites, mostly more homogeneity of crystallites in structure and size and faster coalescence and growth rate as shown in Fig.3. However, there are more some areas showing wide voids and obviously different island height in the case of excessive Ga atoms as revealed in Fig.3 (below).

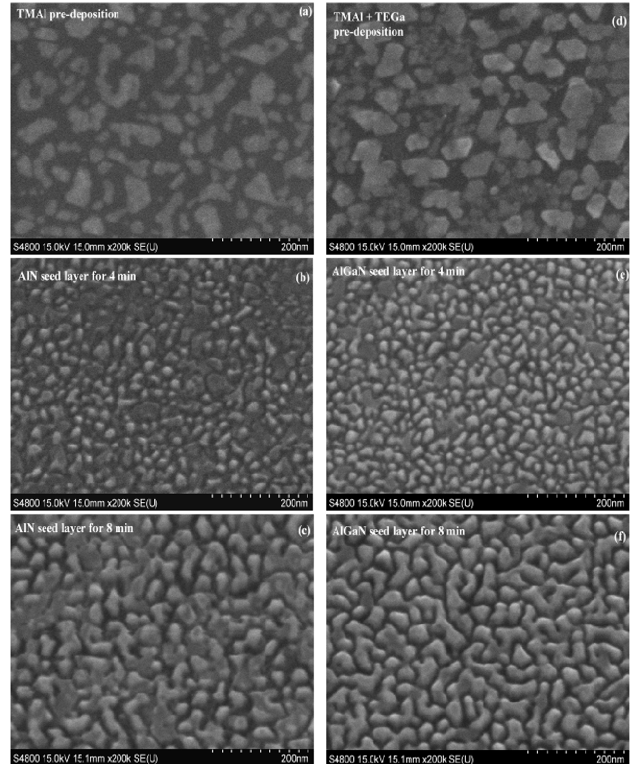

Fig. 2 SEM measurements showing the surface morphology of AlN-nucleation layer without Ga addition (left-hand side), with Ga (right-hand side)

This is because the Ga-atoms have a higher surface diffusion and a higher mobility than Al-atoms do [8]. It clearly refers to lower growth rate and slower coalescence of crystallites by a lower amount of crystallites with likely more spacious areas in the case of no Ga addition as shown in Fig.3 (above). In the case of excessive Ga-addition over the optimized amount, it enables an inferior crystalline quality of subsequent layers due to more different island height and 3D growth rate in some areas.

High-resolution TEM measurements show cross-sectional microstructures of AlN pre-deposition layers with and without Ga in Fig. 4 (above) and 4 (below), respectively. With Ga-addition, more distinguish bonds of Al(Ga)N/Si interface, more periodically crystalline orientation of Al(Ga)N layers, slightly thicker layers and smaller spacious areas as indicated by black arrow in Fig. 4 (below) were observed. These results comparably agree with SEM and AFM measurements that Ga atoms enhance the better-quality nucleation layers with more-uniform crystalline structures, fewer spacious areas and a better homogeneity of crystallite distribution.

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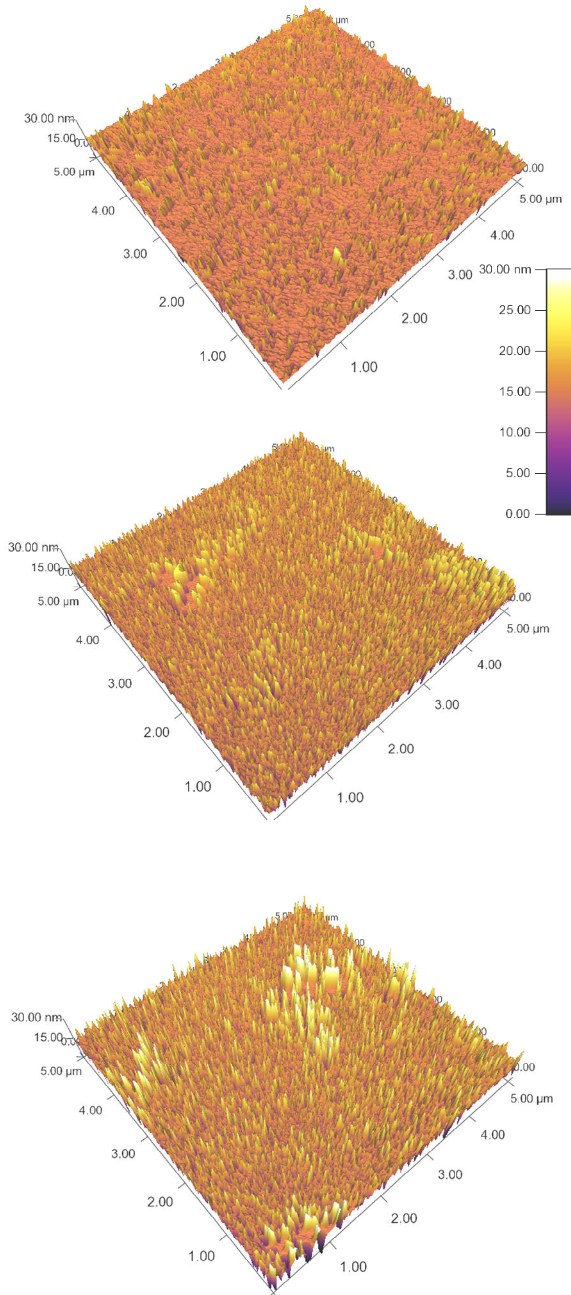

Fig. 3 5×5 μm^2-scan AFM measurements showing the surface morphology of AlN-predeposition layers without Ga addition (above), with TEGa = 40 sccm (middle) and with TEGa = 60 sccm (below)

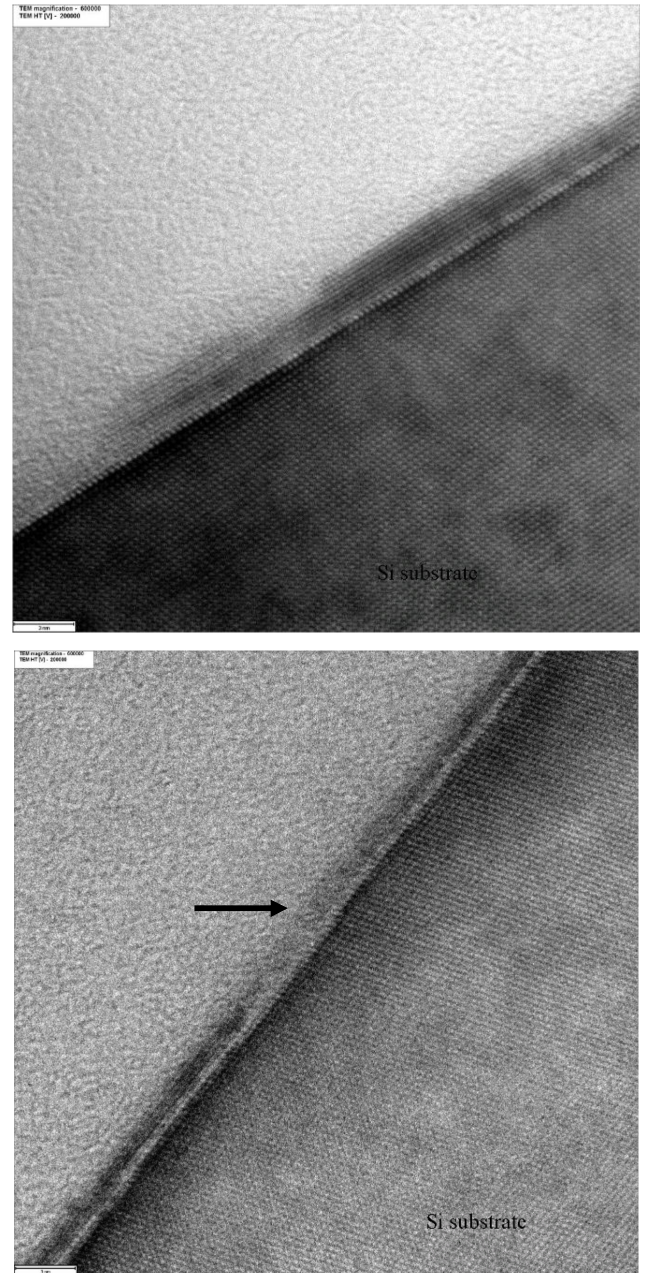

Fig. 4 High-resolution cross-sectional TEM images of Al(Ga)N layers with Ga-addition (above) and without Ga (below) on Si(111) substrates

IV. Conclusion

In summary, optimized Ga-atoms in the Al(Ga)N seed layers play an important role to improve the crystalline quality and reduce the residual strain of subsequent $Al_{0.1}Ga_{0.9}N$ layers by improving surface diffusion to form more-uniform crystallites, a better homogeneity of island distribution, a better crystalline orientation, and then better alignment on the Si-surface. The crystalline quality of the subsequent AlGaN layers grown on Si substrates will be improved and then the high-quality materials to develop the device structures for the UV-range application in further.

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