Paper

Selective Area Growth of III-Nitride and Their Application for Emitting Devices

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ABSTRACT

In this paper, the high quality AlGa_N with high-Al content was grown on in-situ monitoring controlled selective area growth (SAG) GaN and the dislocation density of AlGa_N is $1\text{--}3\times 10^8\text{ cm}^{-2}$. Furthermore, we use SAG to fabricate InGa_N/Ga_N pyramid structures. CL (Cathodoluminescence) measurements reveal that the thickness, CL peak wavelength and CL intensity gradually increased from the bottom to the top of the facet. Furthermore, Vacuum fluorescent display (VFD) based on InGa_N/Ga_N pyramid structures was demonstrated.

KEYWORDS: Selective area growth, AlGa_N, InGa_N/Ga_N pyramid structures Vacuum fluorescent display

1. Introduction

For III-nitride semiconductors, the selective growth (SAG) and the epitaxial lateral overgrowth (ELO) are promising techniques to grow high-quality epitaxial film and to fabricate various semiconductor microstructures such as pyramids, quantum wires, and quantum dots. This structural control technique greatly improve electronic and optoelectronic device perform, for example, prolonging laser diode (LD) lifetime¹⁾, enhancing ultraviolet (UV) light emitting diode (LED) external quantum efficiency²⁾, and decreasing HFETs gate leakage³⁾.

In recent years, SAG and ELO techniques have been well developed as follows: utilization of other masks as tungsten⁴⁾⁵⁾ followed by air-bridge ELO⁶⁾⁷⁾, PENDEO epitaxy⁸⁾ followed by grooved strip structure⁹⁾ and structure substrate²⁾¹⁰⁾¹¹⁾, hetero ELO¹²⁾, Facet Controlled ELO (FACELO)¹³⁾¹⁴⁾, self-organized islands¹⁵⁾¹⁶⁾ followed by nanoscale lateral epitaxial overgrowth (NLEO)¹⁷⁾, and *in situ* SiN nano-mask¹⁸⁾. For Ga_N, these novel structural control techniques can effectively decrease dislocation density and improve crystal quality. However, for other III-nitride semiconductors such as AlGa_N, these structural control techniques don't always affect. The main cause lies on that AlGa_N has smaller lattice constant than Ga_N, which induces tensile stress and easily make film cracking during common SAG and ELO. In addition, Al has highly chemical activity and AlGa_N tend to form polycrystals on the mask. These disadvantages make it difficult to obtain high quality AlGa_N with SAG and ELO.

In this paper, in-situ monitoring controlled SAG is developed to achieve AlGa_N ELO and obtain high quality

AlGa_N with dislocation density of $1\text{--}3\times 10^8\text{ cm}^{-2}$. Meanwhile, SAG is applied to fabricate InGa_N/Ga_N pyramid structures, which is potential to be used to fabricate VFD operated at low-voltage. And the facet dependence of the optical properties of InGa_N is investigated by CL.

2. Experimental

A low-pressure metalorganic vapor phase epitaxy (LP-MOVPE) is used to grow SAG- AlGa_N and InGa_N/Ga_N pyramid structures. (0001) sapphire is used as the substrate. Ammonia (NH₃), trimethylgallium (TMG), trimethylaluminum (TMAI), and trimethylindium (TMI) were used as source materials. Monomethylsilane (CH₃SiH₃) was used as a dopant source to obtain high conductivity.

The growth procedures were as follows: For SAG AlGa_N, a 1.0 μm-thick (0001) AlN/sapphire was used as a substrate. After the deposition of a 100 nm-thick SiO₂ film by RF sputtering on the underlying Si-doped Ga_N, stripe patterns were fabricated by a conventional photolithographic method. The SiO₂ mask had windows with a 7 μm diameter and 3 μm spacing. Then, a Si-doped Ga_N layer was grown at 1080°C at a pressure of 50 Torr. The grow time Growth time is determined by in-situ monitoring. Finally, AlGa_N was grown at 1180°C at a pressure of 60 Torr.

For InGa_N/Ga_N pyramid structures: 3-μm-thick Si-doped Ga_N film was grown on a sapphire (0001) substrate. Then Si-doped Ga_N was patterned with a mask for pyramid structures. Subsequently, Si-doped Ga_N with pyramid structures was grown on the patterned underlayer by MOVPE. Finally, Si-doped InGa_N was

grown at 780 °C with various growth times from 3 min to 27 min.

A high resolution X-ray diffraction (HRXRD) was used to study the crystal quality. And a scanning electron microscope (SEM) with a low-temperature CL setup was used to investigate the optical properties. The CL system provides SEM images, monochromatic CL images and CL wavelength images during one scan.

3. Results and discussion

3.1 AlGaIn grown on in-situ monitoring controlled SAG GaN

Figure 1 is the sketch of SAG AlGaIn: GaN with {11-20} facet was fabricated on the pattern AlN, which was used to SAG AlGaIn. Before GaN coalescent, unmerged GaN layer can reduce dislocation density and release tensile stress; however, after GaN coalescent, GaN had the larger lattice constant than AlGaIn, which induced tensile stress and easily crack AlGaIn. Then, GaN coalescent process has to be controlled during SAG.

For this purpose, an *in-situ* monitoring based on reflecting light intensity is set up to control GaN coalescent process. During GaN growth, the morphology changes from isolated small islands via larger islands to a smooth surface (see Figure 2). Before GaN coalescent, incident light is diffuse reflected on the surface, which cause the reflecting light intensity weak. During GaN coalescent, these isolated small islands merge into larger islands so that reflecting light intensity increases rapidly. At the end of GaN coalescent, mirror reflection is dominated and reflecting light intensity becomes saturation. Then, this in situ monitor can reveal the GaN facet formation and help us to control coalescent process. In our experiments, after 65 min GaN growth, reflecting

light intensity became saturation and the GaN was near to entire coalescent.

Figure 3 is scanning electron microscopy (SEM) images for SAG GaN. The lateral face of stripe was {11-20}, kept about 1~2 μm distance from each other and the height was 3 μm . Here, When $\varphi = 0^\circ$ (the incidence direction of X-ray is parallel to stripe direction), full width at half maximum (FWHM) of (0002) GaN X-ray rocking curves (XRC) was 249 arc sec, and FWHM of (10-12) GaN XRC was 360 arc sec; When $\varphi = 90^\circ$ (the incidence direction of X-ray is perpendicular to the stripe direction), the FWHM value of (0002) GaN XRC was 288 arc sec, and FWHM of (10-12) XRC GaN was 490 arc sec.

After AlGaIn growth, the SEM image is exhibited in Fig.4. The sample exhibited an island-like and smooth morphology. When $\varphi = 0^\circ$, the FWHM of (0002) XRC AlGaIn was 457 arcsec, and FWHM of (10-12) XRC AlGaIn was 1164 arc sec; when $\varphi = 90^\circ$, FWHM of (0002) AlGaIn XRC was 843 arcsec, and FWHM of (10-12) AlGaIn XRC was 1177 arcsec. And the surface CL measurement revealed that dislocation density is $2.9 \times 10^8 \text{ cm}^{-2}$, which was two orders of magnitude lower than that of conventional AlGaIn grown on AlN/sapphire template (Figure 5).

Figure 1 Schematic diagram of AlGaIn ON SAG GaN MODEL

Figure 2 Reflection and temperature profiles for SAG GaN

Figure 3 Cross-sectional and surface SEM images for SAG GaN

Figure 4 Cross-sectional and bird-eyes view SEM images for AlGaIn grown on SAG GaN

Figure 5 CL mapping of AlGaIn grown on flat AlN (up) and SAG-AlN/sapphire (bottom)

3.2 InGaIn on GaN Facets

Using the dot pattern mask, the facet structure of GaN depends on the growth temperature¹⁹⁾²⁰⁾, TMG flow rate¹⁹⁾, and NH_3 partial pressure²⁰⁾. Under the growth condition of a high V/III source gas ratio, a surface that has N-polarity is stabilized, and it becomes more stable at higher pressures or lower temperatures, so that the growth rate tends to decrease, resulting in the $\{1\cdot101\}$ facet. However, under the opposite growth conditions of lower pressures or higher temperatures, the $\{1\cdot101\}$ facet becomes unstable because the surface nitrogen atoms are not stabilized, resulting in the $\{1\cdot100\}$ and $\{0001\}$ facets, which are energetically more favorable. Under our growth condition, the $\{1\cdot101\}$ facet is more stable. Figure 6(a) shows an SEM image of InGaIn on GaN hexagonal pyramid structures. The facet of pyramids had a slope of about 62° with respect to the c-plane corresponding to the $\{10\cdot11\}$ facet. The height of pyramids was about $8\mu\text{m}$. The thickness of InGaIn gradually increased from the lower part to the upper part of the facet. The thickness of the sample with 7 min growth of InGaIn was observed to be

Figure 6 SEM images of InGaIn on GaN facets (a) bird-eyes view image and (b) cross-section image(growth during for 7 min)

Figure 7 SEM images of InGaIn on GaN facets (a) bird-eyes view image and (b) cross-section image(growth during for 7 min)

about 150 nm at the top of the facet but was not measurable in the bottom region, as shown in Figure 6(b).

Figure 7a-c show SEM images, a CL wavelength image and a CL intensity image of the surface for an InGaIn/GaN pyramid structure of the sample with 7 min growth of InGaIn. The GaN underlayer region showed a peak wavelength at about 365 nm, while a peak wavelength at about 450 nm was observed in the region of the InGaIn/GaN pyramid structure. CL spectra of the pyramid structure region shifted to a longer wavelength in the upper part of the facet. The upper part of the facet showed a peak wavelength at 463 nm and the bottom of the facet showed that of 452 nm. In addition, CL intensity gradually increased from the lower part to the upper part of the facet.

Figure 8 shows the dependence of the CL intensity and peak wavelength on InGaIn growth times. The CL intensity was enhanced with increasing InGaIn growth time up to 14 min. With a further increase of the InGaIn growth time, the CL intensity decreased. It could be assumed that the thicker film resulted in a deterioration of the crystal quality.

Figure 9 displays the dependence of the CL peak wavelength on the position from the top to the bottom of the InGaIn facet with different growth times. When the InGaIn growth time was more than 27 min, the distribution of the wavelength from the top to the bottom of the facet was almost constant. No marked, remarkable

Figure 8 Dependence of CL intensity and peak wavelength on InGaN growth time

Figure 9 Dependence of CL peak wavelength on the position from the top to the bottom of the facet with different InGaN growth times

CL peak wavelength shift was observed for the sample with 27 min growth of InGaN.

Here, two possible mechanisms are proposed. One is that gallium and indium atoms diffuse towards the top of the facet. Nishizawa et al. have also reported an emission of InGaN/GaN multi-quantum-well (MQW) structures with the {11-22} facet and found that gallium and indium atoms diffused to the top of the facet²¹. As we know, the In-N bond is longer and weaker than the Ga-N bond, and the indium atom preferentially diffuses to the top. Therefore, the upper part of the facet indicates a longer wavelength. For the sample with 27 min growth of InGaN, the diffusion was further enhanced, and a facet with a slope larger than that of the {10-11} facet appeared in the upper part of the facet with the greater thickness. Therefore, the indium diffusion towards the top of the facet was prevented and no CL peak shift was observed.

The other mechanism is the indium composition pulling effect. Shimizu et al. reported that the indium composition was reduced in a thin InGaN film grown on GaN²². It was suggested that indium atoms are excluded from the InGaN lattice during the early growth stages to reduce the deformation energy of the lattice mismatch. It was assumed that the growth rate at the upper part of the

Figure 10 Sketch of VFD application

facet was faster than that at the bottom because of some source gas flow effects. In this case, the shorter wavelength at the bottom of the facet is due to the indium composition pulling effect. It was believed that the lack of an obvious peak shift for the sample with 27 min growth of InGaN was due to the greatly increased film thickness.

Meanwhile, we offer a prototype of Vacuum Fluorescent Display (VFD) with InGaN/GaN pyramid structures with acceleration voltage of 40-100V and Zn-doped GaN, as shown in Fig.10. The results clearly demonstrate that bright VFD with InGaN/GaN pyramid structures can be operated at voltage as low as 100V.

4. Concluding remark

We develop an in-situ controlled SAG to grow other semiconductor besides GaN. With this technique, we obtain high-quality AlGaIn with dislocation density of $1\text{--}3 \times 10^8 \text{ cm}^{-2}$, which are two orders of magnitude lower than that of conventional AlGaIn. The reduction of TD density is due to strain relaxation and ELO of AlGaIn.

Furthermore, we use SAG to fabricate InGaIn/GaN pyramid structures. For samples with InGaIn growth time less than 27 min, a redshift of the CL peak wavelength and enhancement of the CL peak intensity were observed from the lower part to the upper part of the facet. Perhaps, it can be attribute to indium diffusion mechanism or the indium composition pulling effect. For the sample with 27 min growth of InGaIn, no obvious change in the CL peak wavelength or intensity was observed because of the hampered indium diffusion. This structure is used to fabricate VFD.

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