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

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ABSTRACT

In this paper, the high quality AlGaN with high-Al content was grown on in-situ monitoring controlled selective area growth (SAG) GaN and the dislocation density of AlGaN is $1\times10^8$ cm$^{-2}$. Furthermore, we use SAG to fabricate InGaN/GaN 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 InGaN/GaN pyramid structures was demonstrated.

KEYWORDS: Selective area growth, AlGaN, InGaN/GaN 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 microstructures such as pyramids, quantum wires, and quantum dots. This structural control technique greatly improve electronic and optoelectronic device performance, for example, prolonging laser diode (LD) lifetime, enhancing ultraviolet (UV) light emitting diode (LED) external quantum efficiency, and decreasing HFETs gate leakage.

In recent years, SAG and ELO techniques have been well developed as follows: utilization of other masks as tungsten following by air-bridge ELO, PENDEO epitaxy followed by grooved strip structure and structure substrate, Facet Controlled ELO (FACELO), self-organized islands followed by nanoscale lateral epitaxial overgrowth (NLEO), and in situ SiN nano-mask. For GaN, these novel structural control techniques can effectively decrease dislocation density and improve crystal quality. However, for other III-nitride semiconductors such as AlGaN, these structural control techniques don't always affect. The main cause lies on that AlGaN has smaller lattice constant than GaN, which induces tensile stress and easily make film cracking during common SAG and ELO.

In this paper, in-situ monitoring controlled SAG is developed to achieve AlGaN ELO and obtain high quality AlGaN with dislocation density of $1\times10^8$ cm$^{-2}$. Meanwhile, SAG is applied to fabricate InGaN/GaN pyramid structures, which is potential to be used to fabricate VFD operated at low-voltage. And the facet dependence of the optical properties of InGaN is investigated by CL.

2. Experimental

A low-pressure metalorganic vapor phase epitaxy (LP-MOVPE) is used to grow SAG AlGaN and InGaN/GaN pyramid structures. (0001) sapphire is used as the substrate. Ammonia (NH$_3$), trimethylgallium (TMG), trimethylaluminum (TMA), and trimethylindium (TMI) were used as source materials. Monomethylsilane (CH$_3$SiH$_3$) was used as a dopant source to obtain high conductivity.

The growth procedures were as follows: For SAG AlGaN, a 1.0 µm-thick (0001) AlN/sapphire was used as a substrate. After the deposition of a 100 nm-thick SiO$_2$ film by RF sputtering on the underlying Si-doped GaN, stripe patterns were fabricated by a conventional photolithographic method. The SiO$_2$ mask had windows with a 7 µm diameter and 3 µm spacing. Then, a Si-doped GaN layer was grown at 1080°C at a pressure of 50 Torr. The grow time was determined by in-situ monitoring. Finally, AlGaN was grown at 1180°C at a pressure of 60 Torr.

For InGaN/GaN pyramid structures: 3 µm-thick Si-doped GaN film was grown on a sapphire (0001) substrate. Then Si-doped GaN was patterned with a mask for pyramid structures. Subsequently, Si-doped GaN with pyramid structures was grown on the patterned underlayer by MOVPE. Finally, Si-doped InGaN 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 AlGaN grown on in situ monitoring controlled SAG GaN

Figure 1 is the sketch of SAG AlGaN: GaN with {11-20} facet was fabricated on the pattern AlN, which was used to SAG AlGaN. 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 AlGaN, which induced tensile stress and easily crack AlGaN. 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 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 m distance from each other and the height was 3µm. Here. When φ = 0° (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 φ = 90° (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 AlGaN growth, the SEM image is exhibited in Fig. 4. The sample exhibited an island-like and smooth morphology. When φ = 0°, the FWHM of (0002) XRC AlGaN was 1164 arcsec, and FWHM of (10-12) XRC AlGaN was 1177 arcsec. When φ = 90°, FWHM of (0002) AlGaN XRC was 843 arcsec, and FWHM of (10-12) AlGaN XRC was 1154 arcsec. And the surface CL measurement revealed that dislocation density is 2.9×10^6 cm^-2, which was two orders of magnitude lower than that of conventional AlGaN grown on AlN/sapphire template (Figure 5).
8.2 InGaN on GaN Facets

Using the dot pattern mask, the facet structure of GaN depends on the growth temperature, TMG flow rate, and NH3 partial pressure. Under the growth condition of a high VIII source gas ratio, a surface that has N-polarity is stabilized, and it becomes more stable at lower pressures or lower temperatures, so that the growth rate tends to decrease, resulting in the {1-101} facet. However, under the opposite growth conditions of lower pressures or higher temperatures, the {1-101} facet becomes unstable because the surface nitrogen atoms are not stabilized, resulting in the {1-100} and {0001} facets, which are energetically more favorable. Under our growth condition, the {1-101} facet is more stable. Figure 6(a) shows an SEM image of InGaN on GaN hexagonal pyramid structures. The facet of pyramids had a slope of about 62° with respect to the c-plane corresponding to the {10-11} facet. The height of pyramids was about 8 μm. The thickness of InGaN gradually increased from the lower part to the upper part of the facet. The thickness of the sample with 7 min growth of InGaN was observed to be about 150 nm at the top of the facet but was not measurable in the bottom region, as shown in Figure 6(b).

Figure 7(a–c) show SEM images, a CL wavelength image, and a CL intensity image of the surface for an InGaN/GaN pyramid structure of the sample with 7 min growth of InGaN. The GaN underlayer region showed a peak wavelength at about 370 nm, while a peak wavelength at about 450 nm was observed in the region of the InGaN/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 InGaN growth times. The CL intensity was enhanced with increasing InGaN growth time up to 14 min. With a further increase of the InGaN 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 InGaN facet with different growth times. When the InGaN 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
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 facet. 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 GaN. 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 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 AlGaN with dislocation density of 1-3×10^6 cm^2, which are two orders of magnitude lower than that of conventional AlGaN. The reduction of TD density is due to strain relaxation and ELO of AlGaN.

Furthermore, we use SAG to fabricate InGaN/GaN pyramid structures. For samples with InGaN 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 InGaN, 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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