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Coalescence overgrowth of GaN nano-columns with metalorganic chemical vapor deposition

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Abstract

The authors demonstrate the coalescence overgrowth of GaN nano-columns on a (111) Si substrate with metalorganic chemical vapor deposition to show high-quality optical properties in the overgrown film. Plan-view scanning electron microscopy (SEM) shows coalesced surface morphology, although hexagonal structures are still visible in the images. The cross-section cathodoluminescence (CL) image shows more efficient emission in the overgrowth layer than from the nano-column layer. The plan-view CL image demonstrates that the emitted light is mainly from the hexagonal structures. The photoluminescence measurement result indicates that the emission efficiency of the overgrown layer is even higher than that of an undoped GaN thin film of high quality. The presence of hexagonal structures correlates to surface roughness values in the range of several nanometres.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Given their inherent lateral strain relaxation and potential for reducing strain-induced defects that originate from nucleation layers on lattice-mismatched substrates, GaN nano-columns on sapphire or other substrates have drawn much attention. GaN nano-columns of low threading dislocation densities have been grown with molecular beam epitaxy (MBE) or metalorganic chemical vapor deposition (MOCVD) [1–5]. Normally, the MBE-grown nano-columns are more mutually parallel and vertical to the substrate surface. Such nano-columns can be grown from an AlN nucleation layer [6] or from a patterned

GaN template [7]. In growing such nano-columns, the most important factor is producing an N-rich growth condition, which can be implemented with a low III/V ratio or an enhanced desorption rate of Ga atoms at a high growth temperature. The column cross-sectional size and area density is also controlled by the III/V ratio [4]. In general, a sample with smaller-diameter columns has a higher column density.

Due to current technological limitations, it is difficult to implement device fabrication with a structure of nano-columns. The nanostructures make the formation of metal contacts difficult. Coalescence overgrowth of GaN nano-columns becomes an important procedure in preparing a

GaN template with a low threading dislocation density for subsequent device fabrication. Coalescence overgrowth on GaN or InGaN/GaN nano-columns with MBE has been attempted for fabricating light-emitting diodes [8, 9]. However, the detailed morphology and the optical properties of such an overgrowth layer, particularly that grown with MOCVD, have never been reported. Detailed characterization is crucially important for the subsequent device fabrication. In this paper, we report the morphology and cathodoluminescence (CL) characterization results of a sample of coalescence overgrowth by MOCVD on GaN nano-columns, which are grown by MBE on (111) Si substrate. Recently, GaN growth on Si substrate has become an important topic for research because of its advantages, including low cost, wide availability, high thermal conductivity, and potential for integrating with electronics devices, when compared with sapphire-based growth [10–12]. However, GaN growth on Si substrate suffers from several problems, particularly the wafer crack due to the built strain during growth [13, 14]. The successful coalescence overgrowth of nano-columns provides an alternative method for fabricating high-quality GaN templates on Si substrates. In this research, it is found that the coalesced surface shows residual hexagonal structures with surface roughness in the range of several nanometres. The CL characterization demonstrates stronger luminescence intensity in the overgrowth layer, when compared with that of the nano-columns. The emission efficiency of the overgrowth layer is even stronger than that of a high-quality undoped GaN thin film grown on a sapphire substrate.

2. Sample preparation and measurement conditions

The MBE-grown nano-columns were formed under the following conditions. Before growth, the Si(111) substrate was cleaned with a dip in 10% aqueous HF solution, followed by rinsing in deionized water, before being blown dry. The substrate was heated to approximately 710 °C for the AlN layer. The active nitrogen flux was provided by an EPI Unibulb plasma source operated at 330 W and 2.0 sccm in nitrogen flow rate. Substrate temperatures were measured using an optical pyrometer. Prior to igniting the nitrogen plasma, a small amount of aluminum was deposited onto the silicon surface to prevent the formation of silicon nitride. The AlN nucleation layer was first grown at a growth rate of 0.04 nm s⁻¹ before the deposition of GaN nano-columns. After the AlN nucleation layer was deposited, the substrate temperature was raised to 810 °C for growing GaN nano-columns at a growth rate of 0.07 nm s⁻¹. The nano-column cross-sectional size is about 100 nm with the column density estimated to be 10⁹ cm⁻². The height of the nano-columns is about 2 μm. An MOCVD-overgrown GaN layer of about 2.6 μm in thickness was deposited at 1000 °C in a Veeco P75 system. The growth pressure was 200 Torr and the estimated growth rate was about 0.44 nm s⁻¹. The V/III ratio was 2600 and the flow rate of NH₃ was 1000 sccm. Trimethylgallium was used as Ga precursor. The scanning electron microscopy (SEM) and CL measurements were performed with a JEOL JSM 6700F system. The acceleration voltage of the electron beam was 15 kV. Before the SEM and CL measurements, a 3 nm Pt layer was coated on the sample to minimize the charging effect during the measurements.

Figure 1. Cross-sectional SEM image (a) and CL image (b) of the sample.

The x-ray diffraction (XRD) measurement in the (0002) plane shows a full-width at half-maximum (FWHM) of 3723 arcsec in the rocking curve. This value is much higher than the typical level of <200 arcsec in a high-quality undoped GaN thin film. The large rocking curve FWHM is attributed to the composition of numerous domains of slightly different crystalline orientations within the x-ray beam in the XRD measurement. In other words, in the overgrowth layer, domains of different crystalline orientations originating from different nano-columns or different groups of nano-columns exist.

3. SEM and CL results

Figures 1(a) and (b) show a cross-section SEM image and the corresponding CL image, respectively, of the overgrown sample. In the initial stage of MOCVD overgrowth, nano-columns may merge into larger columns. Therefore, nano-columns under the overgrowth layer in the SEM image show cross-sectional sizes larger than 100 nm. In the SEM image, different columns seem to be coalesced in the overgrowth layer. The top surface is essentially flat and smooth. The CL image in figure 1(b) shows a strong contrast between the bright emission in the overgrowth layer and the dark background in the nano-column layer. The strong emission contrast has two possible causes. First, during the overgrowth stage, low-quality GaN may have filled in the space between columns and dominated the CL behavior. The second possible cause is that the optical

Figure 2. Plan-view SEM image (a) and CL image (b) of the sample.

quality of the higher-temperature GaN overgrowth layer is better than that of the lower-temperature grown GaN nanocolumns. Normally, a lower-temperature grown GaN sample has a higher point defect density, leading to lower internal quantum efficiency. The CL emission through the top surface makes this region quite bright, as shown in figure 1(b).

Figures 2(a) and (b) show a typical plan-view SEM image and its corresponding CL image, respectively. In figure 2(a), hexagonal structures can be seen. Some hexagons have flat top facets of a few microns in dimension. The height of a hexagon decreases from the top facet to the boundary of a domain, which is several microns in size. Different domains look well connected in morphology. The CL image in figure 2(b) also shows quite a strong contrast between the bright and dark regions. The dark regions are always located around the boundaries of the domains. In other words, the optical property near the center of a domain is better than that near its boundary. Because the emission from the nanocolumns is less efficient, the nanocolumns beneath the boundary regions do not contribute much to the CL intensity.

Figure 3 shows the normalized CL spectra from the plan-view (p-CL) and cross-section (c-CL) images at room temperature. One can see weak yellow luminescence in both cross-section CL spectra originating from extended defects [15]. Both CL spectra have peaks around 363 nm with tails on the long-wavelength side. The inset (a) shows

Figure 3. Normalized CL spectra in the plan-view (p-CL, continuous curve) and cross-sectional (c-CL, dashed curve) measurements. Inset (a): normalized PL spectra at 10 K (PL-10 K, continuous curve) and 300 K (PL-300 K, dashed curve); inset (b): PL spectra at 300 K of the overgrowth sample (PL-300 K, dashed curve) and of a high-quality undoped GaN thin-film sample (u-GaN, continuous curve) showing their relative intensities.

the photoluminescence (PL) spectra at 10 and 300 K. The PL spectral peak at 300 K around 364 nm is quite consistent with the CL spectral peak at 363 nm. The major peak of the PL spectrum at 10 K is slightly blue shifted, as expected. In this spectrum, a minor peak on the long-wavelength side at 368 nm can be clearly seen. It is attributed to certain strongly localized defects [16, 17]. In the inset (b) of figure 3, we compare the PL intensities of the overgrown layer and a high-quality 3 μm thick GaN thin film (u-GaN), in which the rocking curve FWHM in the (0002) plane is 190 arcsec and that in the (102) plane is 296 arcsec. To obtain the result in inset (b), the PL measurement setup was carefully arranged such that the relative PL intensity between the two samples could be measured. In this inset, one can see that the PL intensity of the overgrown layer, which is also around 3 μm in thickness, is significantly stronger than that of the u-GaN sample (a factor of 7.08 stronger in integrated PL intensity). Meanwhile, temperature-dependent PL measurement indicates that the internal quantum efficiency of the overgrown layer is 16.67%, while that of the u-GaN sample is only 7.04%. These comparisons lead to the conclusion that the optical quality of the overgrown layer is actually quite good although certain regions are not emissive, as shown in figure 2(b).

4. Discussions and conclusions

As shown in figure 2(a), hexagonal structures exist on the top surface of the overgrown layer. To understand the surface morphology more quantitatively, we scan the sample surface with atomic force microscopy (AFM). Figure 4(a) shows a 7 $\mu\text{m} \times 7 \mu\text{m}$ AFM image demonstrating part of a hexagon. Here, two sides of the hexagon can be clearly seen. Figure 4(b) shows a line-scan profile along the straight line in figure 4(a). The difference in height between the maximum and the minimum, indicated by the two marks, is 14.18 nm. This value represents the largest variation in surface height we could find in more than ten AFM images we took on this sample.

Figure 4. (a) An AFM image showing part of a hexagonal structure and (b) the line-scan profile.

The surface roughness evaluated based on this AFM image is 5.69 nm. The left corner of this AFM image is expected to approach the tip of the hexagon. However, the AFM image shows a descending trend that can be due to the tilt of the hexagon itself.

In summary, we have demonstrated the coalescence overgrowth of GaN nano-columns with MOCVD to show high-quality optical properties. The plan-view SEM image showed essentially coalesced surface morphology although hexagonal structures could still be seen. The cross-section CL image showed more efficient emission from the overgrowth layer than from the nano-columns. The plan-view CL image demonstrated that the emission was mainly from the hexagonal structures. The PL measurements indicated that the emission

efficiency of the overgrown layer was even higher than that of an undoped GaN thin film of high quality. The hexagons led to surface roughness values in the range of several nanometres.

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References

- [1] Yoshizawa M, Kikuchi A, Mori M, Fujita N and Kishino K 1997 *Japan. J. Appl. Phys.* **2** [36 L459](#)
- [2] Tu L W, Hsiao C L, Chi T W, Lo I and Hsieh K Y 2003 *Appl. Phys. Lett.* **82** 1601
- [3] Araki T, Chiba Y, Nobata M, Nishioka Y and Nanishi Y 2000 *J. Cryst. Growth* **209** 368
- [4] Calleja E, Sanchez-Garcia M A, Sanchez F J, Calle F, Naranjo F B, Munoz E, Jahn U and Ploog K 2000 *Phys. Rev. B* **62** 16826
- [5] Suzuki T, Yagi S and Motooka T 2005 *J. Appl. Phys.* **98** 104303
- [6] Van Nostrand J E, Averett K L, Cortez R, Boeckl J, Stutz C E, Sanford N A, Davydov A V and Albrecht J D 2006 *J. Cryst. Growth* **287** 500
- [7] Wang X, Sun X, Fairchild M and Hersee S D 2006 *Appl. Phys. Lett.* **89** 233115
- [8] Kikuchi A, Kawai M, Tada M and Kishino K 2004 *Japan. J. Appl. Phys.* **43** L1524
- [9] Kusakabe K, Kikuchi A and Kishino K 2001 *Japan. J. Appl. Phys.* **2** [40 L192](#)
- [10] Marchand H, Zhao L, Zhang N, Moran B, Coffie R, Mishra U K, Speck J S, DenBaars S P and Freitas J A 2001 *J. Appl. Phys.* **89** 7846
- [11] Dadgar A *et al* 2003 *J. Cryst. Growth* **248** 556
- [12] Dadgar A *et al* 2007 *Thin Solid Films* **515** 4356
- [13] Nikishin S A, Faleev N N, Antipov V G, Francoeur S, Grave de Peralta L, Seryogin G A, Temkin H, Prokofyeva T I, Holtz M and Chu S N G 1999 *Appl. Phys. Lett.* **75** 2073
- [14] Wu J, Han X, Li J, Li D, Lu Y, Wei H, Cong G, Liu X, Zhu Q and Wang Z 2005 *J. Cryst. Growth* **279** 335
- [15] Li G, Chua S J, Xu S J, Wang W, Li P, Beaumont B and Gibart P 1999 *Appl. Phys. Lett.* **74** 2821
- [16] Rieger W, Dimitrov R, Brunner D, Rohrer E, Ambacher O and Stutzmann M 1996 *Phys. Rev. B* **54** 17596
- [17] Wetzel C, Fischer S, Kruger J, Haller E E, Molnar R J, Moustakas T D, Mokhov E N and Baranov P G 1996 *Appl. Phys. Lett.* **68** 2556