Epitaxial overgrowth of GaN nanorods on Si (111) substrates by rfplasma-assisted molecular-beam epitaxy

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1. Introduction

III-nitride semiconductors are promising materials with great potential for high-power device and optoelectronic applications. Because of the lack of suitable substrate, most GaN layers have been grown on sapphire (0001) substrates, which have some disadvantages, such as unavailability of large sizes and insulating property. To avoid these disadvantages, the growth of GaN on Si (111) has attracted considerable academic and commercial interest in recent years. Si substrates are lower cost, larger sizes, higher crystalline quality and have potential for mature device processing and device integration with Si circuitry. However, growing device-quality GaN epilayers on Si substrates is challenging because of the large differences in the lattice constant (20%) and thermal expansion coefficient (56%) between GaN and Si. High dislocation density of 10⁸-10¹⁰ cm⁻², stress induced piezo-polarization field and cracks, which were found in the thick GaN epilayers grown on Si, degrade the electrical and optical properties of the devices. On the other hand, at nitrogen rich condition, a reduced Ga surface diffusion results three-dimension (3D) growth and formation of GaN nano-rods (NRs) [1]. GaN NRs are hexagonal-column single crystals, which are selforganized and perpendicular to substrates which have better crystal quality than that of the epilayers directly grown on Si [2]. The GaN NRs are typically 20-200nm in diameter and 0.2-2.0 μ m in height with a density of 0.2-1.8×10¹⁰ cm⁻². It has been revealed that GaN nanorods have highly efficient photoluminescence (PL) due to low dislocation density [3]. Recently, Kusakabe et al. found that GaN layer overgrown on GaN NRs has much better crystal and optical quality than that of GaN layer directly grown on Si [4].

In this study, GaN epilayer were overgrown with different size of GaN NRs on Si (111) substrates by RF-MBE. The effects of surface morphology and optical properties of the overgrowth layer were investigated. The surface morphology of overgrown GaN was investigated by scanning electron microscopy (SEM). The optical properties were evaluated by photoluminescence (PL) measurements at 13 K was performed with a 325nm He-Cd laser as an excitation source.

2. Experimental

Overgrown GaN films on GaN NRs investigated in this study were grown by ULVAC RF-plasma molecular beam epitaxy (MBE) system. The 2-inch Si (111) wafers (p-type doping) were chemically cleaned by 10% HF without rinsing in DI water to suppress oxide formation. A clear Si (7×7) surface reconstruction was obtained by reflection high-energy electron diffraction (RHEED) at a substrate temperature of 830°C before the deposition of GaN NRs. The GaN NRs were directly grown on Si (111) substrates at 900°C without AlN nucleation layer under various Ga flux (Φ_{Ga}) conditions to obtain NRs of different sizes. Φ_{Ga} was adjusted to be 7.1×10^{-8} , 2.0×10^{-7} and 3.6×10^{-7} ⁷ torr for rod diameter of 45, 95, and 125 nm, respectively. During the growth, the nitrogen flow rate and RF input power were maintained at 4.0 sccm and 500 W, respectively. Followed the growth of nano-rods, Φ_{Ga} was ramped up to 7.12×10^{-7} torr to start the GaN epitaxial overgrowth. For comparison, GaN layer is also grown on the AlN buffer layer at the same condition as the GaN overgrowth layer.

3. Results and Discussion

Figure 1 shows the RHEED patterns of (a) GaN NRs and (b) GaN epitaxial overgrowth on the NRs with the incident electron beam along $\langle 10\bar{1}0 \rangle$ azimuth. RHEED analysis directly verified that the NRs were wurtzite single crystals structure with same growth orientation as the Si (111) substrate. In contrast with the spotty RHEED pattern observed for the GaN nano-rods (Fig.1 (a)), the streaky pattern in (Fig.1(b)) implies two-dimensional (2D) layer structure of the GaN overgrowth.



Fig. 1 RHEED patterns of (a) GaN nano-rods and (b) GaN overgrown thin film on GaN NRs along $\langle 10\bar{1}0 \rangle$ azimuth.

Figure 2 shows the side-view and tilted top-view of SEM images for GaN epitaxial overgrowth on the NRs and GaN epilayer on AlN buffer. In Fig. 2(a), GaN overgrown on small and dense NRs results vertical growth and

formation of separated and larger NRs. As the diameters of NRs were increased, the lateral growth was enhanced. As a result, two-dimensional overgrowth layer can be observed in the SEM image. The further increasing diameter of NRs results in the flat surface morphology as shown in Fig. 2(c), which is comparable to that of GaN directly grown on Si(111) substrate as shown in Fig. 2 (d).



Fig. 2 Side-view (left) and tilted top-view (right) SEM images of GaN overgrowth on NRs of diameters 45 nm (a) , 95 nm (b) and 125 nm (c) and GaN thick layer on AlN buffer (d).

Figure 3 (a) shows the typical low temperature PL spectra of GaN epitaxial overgrowth on the NRs structure with different size, and (b) LT-PL peak energy and full width at half maximum (FWHM) as a function of Ga flux. In Fig. 3 (a), the dominant PL peak is attributed to the donor-bound exciton emission. The weaker PL structure is assigned to the stacking fault emission [5]. The PL peak energies of the overgrowth GaN epilayer on the 45, 95, and 125 nm NRs are 3.485eV, 3.479eV, and 3.471 eV, respectively. The decreasing PL peak energy implies the strain relaxation [4, 6]. For the overgrowth GaN epilayer on the 125 nm NRs, the PL emission energy is very close to the bulk GaN. It indicates that the overgrowth GaN epilayer is nearly strain free.



Fig. 3 (a) Low-temperature PL spectra of GaN epitaxial overgrowth on the NRs structure with different size, and (b) LT-PL peak energy and FWHM of the peaks as a function of Ga flux.

4. Conclusion

The growth behavior of GaN layers overgrown on NRs on Si substrates by RF-plasma MBE was discussed. The coalescence of GaN overgrowth on larger GaN NRs results in the formation of two dimensional epitaxial layer. The PL spectra show that the GaN films overgrowth on lagre size NRs is nearly strain free.

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