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Effect of hexagonal hillock on luminescence characteristic of multiple quantum wells structure

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ABSTRACT

GaN based ultraviolet multiple quantum well structures grown on a *c*-plane sapphire substrate by metal organic chemical deposition showed a microstructure with a large amount of huge hexagonal hillocks. The polarity of the sample is confirmed by etching with sodium hydroxide solution. The luminous intensity distribution of a typical hexagonal hillock was investigated by the photoluminescent mapping and the luminous intensity at hillock top regions was found to be 15 times higher than that of the regions around hillocks. The reduction of dislocations, the decreasing of the quantum confirmed stack effect caused by semipolar plane and the inclination of the sidewalls of the hexagonal hillock were responsible for the enhancement of luminous intensity.

1. Introduction

In recent years, III-nitrides semiconductor materials have been widely used in the commercial production of high-power electronic devices [1], high-brightness light-emitting diodes [2–4] and room temperature laser diodes [5]. As a typical wide bandgap semiconductor material, GaN has great potential of applications due to its outstanding properties. GaN grown on a *c*-plane sapphire substrate has a hexagonal crystal structure showing either Ga-polarity or N-polarity, which has great influence on the growth behaviors, surface morphologies and electrical and optical properties [6–8]. Both N-face and Ga-face GaN structures contain a lot of hexagonal hillocks that caused the degradation of crystal quality and electrical characteristic [9–12]. In general, the existence of hexagonal hillocks on GaN surface has limited the development of GaN devices. Theoretical and experimental studies have focused on structural characteristic such as stress and dislocations, or electrical characteristics of hexagonal hillocks [9,13–15]. For example, according to a study of Marini et al., reductions in hexagonal density have been achieved by optimizing the high temperature AlN nucleation layer and using an indium surfactant in GaN overgrowth [14]. Many authors have focused on reducing hexagonal hillocks and improving crystal quality or ameliorating electrical characteristic. However, to our knowledge, the influence of hexagonal hillocks on the optical characteristics of multiple quantum well (MQW) structures has rarely been examined.

In this work, we mainly focus on the optical characteristics of hexagonal hillocks in GaN based ultraviolet MQW structures on a *c*-plane sapphire. Room temperature photoluminescence (PL) mapping were used to study the optical properties. The PL result indicates that luminous intensities of hexagonal hillocks were 15 times higher than areas without them. The reasons of this enhancement were speculated to be reduced dislocations, decreased quantum-confined stark effect (QCSE) caused by semipolar plane, and inclined hillock sidewalls.

2. Experimental

GaN based ultraviolet MQW structures were grown on a *c*-plane sapphire substrate by metal organic chemical deposition system. Trimethylgallium and ammonia precursors were used as Ga and N sources, respectively, and H_2 was used as carrier gas. A 3-μm-thick GaN buffer layer was grown at the temperature of 1080 °C on the *c*-plane sapphire substrate with 25-nm-thick AlN sputtered layer followed by the growth of series of n-type layers, which consisted

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of a 900-nm-thick highly doped *n*-type GaN layer, 120-nm-thick *n*-type AlGaN defect barrier layer and 900-nm-thick n^{++} type electrode contact layer. Then, an active layer consisting of 8 pairs of $In_{0.06}Ga_{0.94}N$ (1.5 nm)/GaN (11 nm) MQWs were deposited on those *n*-type layers, followed a 20-nm-thick AlGaN electrons barrier layer, 180-nm-thick *p*-type GaN layer, and 20-nm-thick p^{++} type GaN layer. After the growth process, the surface morphology of the sample was measured by Scanning Electron Microscope (SEM) (JSM 6700F) and Atomic Force Microscope (AFM) (Dimension Icon). The spatial distributions of dislocations in different surface regions were investigated by cathodoluminescence (CL) spectroscopy. PL measurements were performed to investigate a sample's luminescent intensities and the full width of half maximum (FWHM) of hexagonal hillocks. All PL spectra were measured at room temperature by using a confocal Jobin Yvon LavRam HR800 micro-Raman spectrometer with a charge-coupled device detector and an optical microscopy system. The wavelength of the excitation laser used in this study was 325 nm and the excitation time was 0.1s. The laser power was about 2 mW. The surface polarity of the sample was confirmed by etching with a 10% sodium hydroxide solution at the temperature of 80 °C for 20 min.

3. Result and discussion

Fig. 1 shows the SEM image of the sample grown on sapphire substrate. A large number of hexagonal hillocks on the surface were observed. The diameter of hexagonal hillock is ranging from 70 μm to 100 μm. At the beginning of growth, the large flux of ammonia is an indispensable reason to promote some of the surface to form hexagonal hillock structures. Formation of these structures is probably related to substrate surface conditions and very limited surface diffusion for Ga atoms on N-face surface [15].

Sample polarity needed to be identified before studying the optical properties. Based on the very different chemical reactivity of N-face and Ga-face GaN to sodium hydroxide solution, polarity was analyzed by hot wet alkaline etching [16]. For the N-polarity film, sodium hydroxide solution etches the film very quickly, resulting in a drastic change of surface morphology. In contrast, the surface of a Ga-face sample is unaffected by etching [17]. As shown in Fig. 2, SEM image indicates that the sample surface after etching is nearly identical to pre-etching images (Fig. 1). This result indicates that the polarity of the sample is Ga-face.

The microstructure of these hexagonal hillocks was further investigated by AFM. It can be found that the sidewall of the hexagonal hillock during the growth process has an inclined plane as

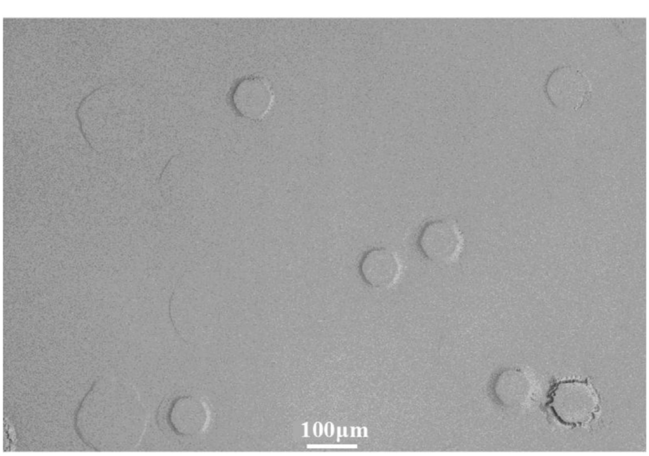

Fig. 1. SEM image of hexagonal hillocks on GaN surface.

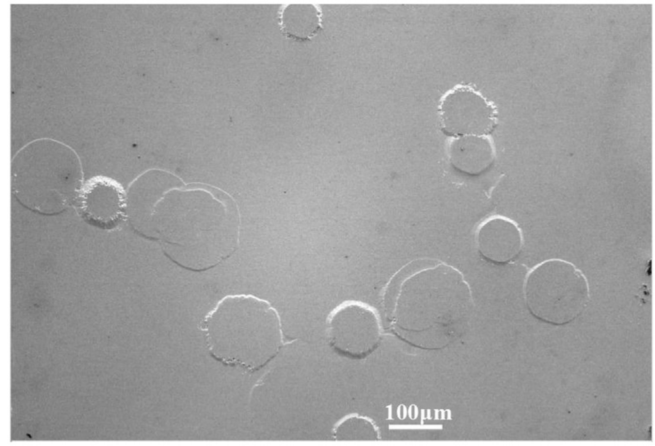

Fig. 2. SEM image of hexagonal hillocks on GaN after surface etching.

shown in Fig. 3. Those AFM images can clearly indicate that the sidewall of the hexagonal hillocks possess a certain angle. In contrast to polar orientation, the sidewall of the hexagonal hillock shows a semipolar orientation [18].

Comparison of the photoluminescent intensity of hexagonal hillocks and flat regions shows that the PL intensity at hillock tops is almost 15-fold higher than that in flat regions lacking hillocks (Fig. 4). Those two regions have the peak wavelengths of 393 nm and 386 nm, respectively, which were both in the ultraviolet range. This observed red shift in hillocks versus flat regions is speculated to be caused by the higher Indium incorporation efficiency in semipolar regions [19,20]. These PL results indicate that hexagonal hillocks appear to significantly enhance the luminous intensity.

In order to further investigate the optical characteristics of hexagonal hillocks in GaN based MQW structures, PL mapping measurements were carried out. Fig. 5 illustrates the intensity and FWHM maps of the hexagonal hillock in a $100 \times 100\,\mu m^2$ zone measured in the surface of GaN, and the dashed lines in Fig. 5 show the shape of the hexagonal hillock. The mapping data is given by the false-color images with the hues ranging from yellow for the largest values to black for the smallest values. Individual PL value is fitted to a Lorentzian-Guassian mixed function to extract values for the intensities and FWHMs.

The intensities in the center of the hexagonal hillock are larger than other regions in Fig. 5(a), and gradually decrease from the center to the region outside the hillock. At the same time, the variations of the FWHM exhibit opposite trend. The top region of a hexagonal hillock possesses the smallest FWHMs, while the sidewalls have larger FWHM values, which is illustrated in Fig. 5(b). It is well known that the intensity and the FWHM of the PL mapping can reflect the crystal quality of GaN films [21]. Generally, the dislocations act as nonradiative recombination centers [22], and the smaller FWHM value indicates the lower dislocation density and the better crystal quality. Therefore, the top region of the hexagonal hillock on the surface possessed the lowest dislocation density [13].

Cathodoluminescence (CL) studies were carried out to further identify the spatial distributions of dislocations in different surface regions. Fig. 6 shows the CL mapping results of the top and surrounding regions of a hillock. It can be clearly noticed that the number of dark spots of the top region of hillock is smaller than that of the region outside the hillock. The difference of dislocation distribution is in agreement with the luminous intensity observed in PL mapping measurements, which further confirms that reduced dislocation density enhanced the luminous intensity.

It is well known that when GaN films are grown along polar