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Efficient broad color luminescence from InGaN/GaN single quantum-well nanocolumn crystals on Si (111) substrate

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ABSTRACT

Nanocolumn InGaN/GaN single quantum well crystals were deposited on Si (111) substrate with nitrified Ga dots as buffer layer. Transmission electron microscopy image shows the crystals' diameter of 100–130 nm and length of about 900 nm. Nanoscale spatial phase separation of cubic and hexagonal GaN was observed by selective area electron diffraction on the quantum well layer. Raman spectrum of the quantum well crystals proved that the crystals were fully relaxed. Room temperature photoluminescence from 450 to 750 nm and full width at half maximum of about 420 meV indicate broad color luminescence covering blue, green, yellow and red emission, which is helpful for the fabrication of tunable optoelectronic devices and colorful light emitting diodes.

1. Introduction

GaN-based light emitting diodes (LEDs) have been attractive for several solid state lighting (SSL) applications, such as display technology, back lighting, light sources for general illumination and et al. LEDs based on III-nitride low-dimensional structures are capable of light emission from the near-ultraviolet (NUV) to the red. InGaN/GaN quantum well (QW) as active layer is an important structure for the above applications. By modulating the In or Al concentration in the GaN layer, wide-range emission can be obtained. Conventional GaN-based LEDs usually emit nearly monochromatic output light. This is very helpful for the fabrication of display devices. However, it is not favorable to solid state white lighting since monochromatic light degrades the color-rending properties. The most conventional methods to achieve white LEDs are the use of color mixing of a NUV InGaN LEDs and a bule/green/red phosphor [1], RGB LEDs [2], or a blue LED and a yellow phosphor [2,3]. Nevertheless, phosphor-based white light emitters have several disadvantages such as stokes shift energy loss, non-radiative and optical losses, relatively short life-time of the phosphors [4,5]. For these reasons, a lot of effort has been devoted to obtaining white emission from a monolithic full-color GaN-based LED (monolithic system without covering any phosphors). The reports on monolithic broad color luminescence GaN-based LEDs such as white lighting and pastels have been based on: InGaN/GaN multiquantum well (MQW) structures [6–8], In-rich InAlGaN/InGaN heterostructures [9], InGaN/GaN quantum dot (QD) structures [10], and doped InGaN/GaN structures [11]. Few studies have focused on that of the InGaN/GaN single quantum-well (SQW) nanocolumn structures.

On the other hand, the combination of GaN with Si is also very interesting, since Si is the most widely used semiconductor and the microfabrication of Si microstructure is easy to implement compared with other semiconductor microstructure. Although the large difference of lattice constant and thermal coefficient between GaN and Si makes it difficult to deposit good quality GaN film on Si substrate, effective methods (such as lateral overgrowth, patterned substrate and nanocolumn growth) were proved to improve the crystal quality on Si substrate [12–17]. It should be noted that Kishino et al. [14,18–20] and Calleja et al. [21–23] via plasma-assisted molecular-beam epitaxy (PA-MBE) to achieve high-quality GaN nanocolumn growth on Si substrates. They found that nanocolumnar heterostructures can be grown on Si (111), Si (100) with AlN [19], GaN [20,23] or AlN/GaN superlattice [18] buffer layer, even on bare Si single-crystal substrates without any buffer layer [21,22]. Specifically, the emission spectra from their InGaN nanocolumns also can be controlled well with adequate structure

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tailoring (process parameter control) or/and selective area growth (SAG) [18,20,23,24]. As for wide-range emission, Lee et al. presented full-color LEDs from non-planar InGaN/GaN MQWs grown on GaN template with truncated hexagonal pyramids [6]. Funato et al. reported InGaN/GaN MQWs grown on *c*-plane (0001) and semipolar {1122} and {1120} microfacets [25]. Shon et al. showed the full-color InGaN-based LEDs on amorphous substrates [26]. However, the broad color LEDs fabricated on these substrates with multiple micro-facets or nanostructures make the materials growth conditions and fabrication process complicated. In this work, we simply used flat Si (111) as substrate and deposited broad color luminescence InGaN/GaN SQW nanocolumn crystals by conventional molecular beam epitaxy (MBE) method.

2. Experimental procedure

Riber-32 molecular beam epitaxy system was used for crystal growth. Single crystal Si (111) wafer was used as substrate. Radio frequency (RF) nitrogen plasma, pure Ga and In were used as N, Ga and In gas source, respectively. The substrate was firstly cleaned by standard RCA method. Then the substrate was transferred into a small high vacuum chamber and heated at a temperature of 200 °C for 12 h to evaporate the absorbed gas from the Si wafer. Secondly, the Si substrate was transferred to the growth chamber and cleaned at a temperature of 805 °C for 25 min to remove the thin oxide layer in the surface. Ga dots were deposited at a temperature of 400 °C for 1 min. Nitridation of the Ga dots was processed when increasing the substrate temperature from 400 °C to 780 °C. High-temperature GaN was deposited at the temperature of 780 °C. Single InGaN/GaN quantum well structure was deposited on the high-temperature GaN layer at a temperature of 525 °C. The thickness of the InGaN well layer is about 5 nm. Finally, 40 nm GaN caplayer was deposited on the quantum well structure.

Surface scanning electron microscopy (SEM) and transmission electron microscopy (TEM) were used to observe the surface and cross-sectional microstructure of the InGaN/GaN film. Raman spectrum of the crystals was measured with a 532 nm laser source at room temperature. Temperature dependent photoluminescence (PL) was analyzed using a He-Cd laser (325 nm, 60 mW) as the excitation source. Optical microscopy image of the PL distribution of the crystals was obtained with a Hg lamp used for the excitation light source. A 405 nm HgI (3S1-3P0) line filter was used to filter the Hg lamp light source. As for the collection of the detection, a high-pass filter of wavelengths longer than 450 nm was used to detect the emission. Reflectivity of the thin film at room temperature was measured with the incident light wavelength from 450 nm to 790 nm.

3. Results and discussion

Fig. 1 shows the SEM images of the InGaN/GaN QW thin film. Fig. 1(a) shows the surface SEM image of the film. It can be easily observed that the thin film exhibits spherical surface morphology. The diameter of the crystal is between 100 and 130 nm. And the InGaN/GaN QW crystals are densely connected with each other. Fig. 1(b) shows the SEM image of the film with the incident electron beam a little tilt from the vertical axis. We can see that the film shows obvious particle microstructure and the rough surface.

The cross-sectional microstructure of the InGaN/GaN film was observed by TEM. Fig. 2 shows the TEM images and the electron diffraction properties of the film. It can be seen from Fig. 2(a) that the GaN crystals are nanocolumn-crystallized. The length of the crystals is about 900 nm and the diameter is about 100–130 nm. Besides, it can be found that the top QW and caplayer part marked with circle is a little larger than the low part of the crystals. Inset of Fig. 2(a) indicates the top part is separated to two columns.

High resolution TEM image is shown in Fig. 2(b). Region A and B correspond to the GaN caplayer and region C corresponds to the GaN underlayer. The well layer between the caplayer and underlayer has a thickness of about 5 nm. A high density of stacking defects can be easily observed in the well layer. Fig. 2(c) and (d) show the selected area electron diffraction (SAED) results of regions in Fig. 2(b). The SAED pattern taken along the $[1\bar{1}00]$ zone axis obtained from the region B, as shown in Fig. 2(c), demonstrates a pure hexagonal wurtzite structure (h-GaN). On the other hand, the SAED pattern obtained from region A in Fig. 2(d) demonstrates a mixed hexagonal/cubic phase. The schematic representation of hexagonal and cubic (c-GaN) phase diffraction spots are shown in Fig. 2(e) and (f), respectively. The results indicate that the nanoscale spatial phase separated h-GaN and c-GaN coexist on the top GaN caplayer and the relationship between them is {111}//{0002} and $[1\bar{1}0]//[11\bar{2}0]$. Spatial phase separation is easy to occur on the nanoscale facet. S. C. Lee et al. used V-groove Si substrate to grow GaN thin film and observed spatial phase separation of GaN on the nanoscale faceted Si substrate [27]. S. Sanorpim et al. also found that the coexistence of cubic and hexagonal structures within a certain growth temperature range [28]. The SAED pattern at the region C also further shows that the GaN layer (it has a wurtzite crystal structure) is epitaxially grown on the Si (111) substrate and that the crystallographic relationship between them is GaN(0002)//Si(111).

Raman spectra of the crystals were measured in the backscattering configuration with a 532 nm laser source at room temperature. In our sample, GaN crystals whose Z-axis (parallel to the hexagonal *c*-axis) is perpendicular to the film layer. As shown in Fig. 3, there are three peaks can be found in Raman spectra for the configurations$Z(--)\bar{Z}$. It is well known that the 520.7 cm^{-1} peak is from the Si substrate. The GaN crystals show two peak positions. One peak position is 567.3 cm^{-1} E_2 mode. The intrinsic peak position of the bulk GaN is reported to be 567.7 ± 0.5 cm^{-1} [29]. Our sample's Raman spectrum peak position is in the region of the intrinsic bulk GaN, which indicates that the GaN crystals are fully relaxed. In addition, the full width at half maximum (FWHM) of peak 567.3 cm^{-1} is 5.91 cm^{-1}. The anther peak at 730.6 cm^{-1} may be attributed to the LO branches of the A_1 mode. With the above configurations, the A_1 (TO) mode and the E_1 mode (both TO and LO) are forbidden in backscattering configuration. Allowed are the LO branch of the A_1 mode and the E_2 mode [30,31]. It should be noted that, although the column is fully relaxed, high density of defects is observed from the TEM image of the column crystals, especially in the well layer. From this point, we can conclude that the full relaxation could not guarantee the crystals free from defects.

Fig. 4 shows optical properties of the InGaN/GaN QW sample dependent on the measurement temperature. Fig. 4(a) shows PL spectra dependent on the measurement temperature. It can be seen that room-temperature PL wavelength varies from about 450 nm to 750 nm, which includes blue, green, yellow, red and part of infrared emission region. Three main peak positions are clearly demonstrated in Fig. 4(a). The three peak positions at room temperature are 516 nm, 599 nm and 669 nm, which respectively shift to 507 nm, 596 nm and 665 nm at a low temperature of 15 K. Wide-range wavelength PL and multiple peak positions are usually due to the phase segregation or concentration fluctuation in the InGaN well layer. V. Perz-solorzano et al. fabricated pyramidal InGaN microstructure which includes a quantum well on the side-walls, a quantum wire at its edges and a quantum dot on the top of the pyramid [32]. Similar three-peak PL curves were also obtained from this pyramid.

Fig. 4(b) shows the FWHM dependent on the measurement temperature. It can be seen that as the measurement temperature