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Effect of the Conduction Type of Si (111) Substrates on the Performance of GaN MQW LED Epitaxial Films

Changda Zheng[1,a], Chunlan MO[1,], Wenqing Fang[1], Fengyi Jiang[1]

[1]National Engineering Technology Research Center for LED on Si Substrate, Nanchang University, 330047, China

[a]zhengchangda@ncu.edu.cn

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Abstract. The present paper prepared a structural epitaxial film of gallium nitride multiple-quantum-well (GaN MQW) blue light-emitting diode (LED) on Si (111) substrates with different conduction types using the metal-organic chemical vapor deposition method. The method prevented the interdiffusion of GaN and Si to achieve high-quality film growth by introducing aluminium nitride (AlN)-interposed layer and rich-Gallium GaN layer with low V/III ratio double buffer layers. Surface analysis shows that the GaN LED epitaxial film on the Si (111) substrate with different conduction types presented an entirely different appearance. The surface roughness of all the samples was less than 3 nm. A much smoother surface of the epitaxial film on the N-type substrate had less roughness, whereas a layered stack surface of the epitaxial film on the P-type substrate had larger surface roughness. The full width at half maximum of the XRD Omega rocking curve with (002) and (102) planes of GaN film grown on the N-type substrate was less than that of the GaN film on the P-type substrate. Furthermore, the film was superior to the samples on the P-type substrate in terms of crystal quality. There was little difference in the peak position of the PL of the epitaxial film on the N-type substrate, but the peak position of the PL of the epitaxial film on the P-type substrate was long and had a large half-peak width. The tensile stress of the GaN film on the P-type substrate was higher. The above results show that the N-type Si (111) substrate with high resistivity is more suitable for the growth of GaN MQW LED epitaxial film.

Introduction

Nitride semiconductor materials are widely used in display, data storage, high frequency and high-power devices, ultraviolet photodetectors, and other related devices due to their excellent characteristics. The poor quality and lack large size of gallium nitride (GaN) single crystal substrate materials has made technologies widely using sapphire and SiC as substrate for hetero-epitaxial GaN materials. Silicon (Si) substrate has some excellent characteristics: cheap, good heat and electrical conductivity, low hardness, made out of large high-quality single crystal materials, and easy to process due to a mature processing device technology. Furthermore, Si used as substrates may achieve optoelectronic integration of silica-based material, causing Si to become one of the most attractive substrate materials for GaN epitaxy [1-3]. However, there is a serious lattice mismatch (17%) and thermal mismatch (>100%) between GaN and Si. Thus, directly growing high-quality GaN epitaxy on Si is difficult.

Researchers have made many attempts to address the above issues, such as adopting the lateral epitaxial method to improve crystal quality, introducing different interlayers to reduce stress. In 1993, Amona et al. [4] first reported the growth of a layer of aluminium nitride (AlN) between high-temperature GaN and Si. They confirmed that low-temperature AIN interlayer reduces the stress received by high-temperature GaN and improves the crystal quality of the epitaxial layer [5,6]. The growths of GaN epitaxial layers without crack on Si (111) substrate were successively reported by adopting the AlN/AlGaN interlayer [7-10]. In 1998, Guha et al. first reported the MBE method to be used to make a double heterojunction luminous diode on an epitaxial wafer with cracks grown on the Si substrate [11]. Tran et al. first produced the blue luminous diode with the epitaxial structure of

InGaN/GaN MQW on Si (111) [12]. Many research groups focused on optimizing growing conditions, such as category, thickness, and interlayer temperature, to improve the characteristics of GaN multiple-quantum-well light-emitting diodes (MQW LEDs). They discussed the effect of annealing on the performance of LED [13,14]. Many research groups had reported that the epitaxial high-quality GaN blue LED structure could be achieved on a Si substrate and a blue LED device when a vertical chip structure is produced [9,15]. The present paper mainly probes the effects of the Si (111) substrate with different conduction types on the performance of the epitaxial layer of GaN MQW LEDs. With AlN and GaN layer with low V/III ratio considered as the interlayer and buffer layer, respectively, high-quality GaN MQW LED epitaxial film without crack, which meets the requirements for producing devices, was grown on Si (111) substrate with different conduction types. The performance of the epitaxial layer was examined using an interference microscope, atomic force microscope (AFM), double crystal X-ray diffraction (DCXRD), and photoluminescence spectrum (PL).

Experimental Details

The experiment was conducted in a Thomas Swan close-coupled showerhead (CCS) MOCVD with Si (111) as substrate. Trimethyl aluminium (TMAl) was used as the Al source, trimethyl gallium (TMGa) as the Ga source, trimethyl indium (TMIn) as the In source, and high purity ammonia (NH3) as the nitrogen source. Silicone hydride (SiH4) and CP2Mg were used as doping agents for the N-type and P-type GaN, respectively. First, the substrate was performed chemical cleaning: the substrate was first immersed in a H2SO4:H2O solution and then underwent acid corrosion with 2% HF. Finally, the substrate was cleaned with de-ionized water and dried by nitrogen flow. After placing inside the MOCVD reaction tube, the substrate underwent high-temperature heat treatment in situ under H2 ambience to remove the surface oxide layer. low-temperature growth AlN interlayer, high-temperature growth 200 nm GaN layer with low V/ III ratio not intentionally doped, 0.4 um GaN layer not intentionally doped, and 2 um N-type GaN layer with doped Si were deposited. Then $In_{0.2}Ga_{0.8}N/In_{0.02}Ga_{0.98}N$ quantum well with five folds was grown under N2 ambience. The growing temperatures of the well and barrier were 720˚Cand 780˚C, respectively, with a thickness of 3 and 9 nm, respectively. Finally, a 0.2um P-type GaN layer doped with Mg was grown in H2 ambience at a temperature of 990˚C [9]. Three kinds of Si substrates were used in the present experiment: N-type Si with electric resistivity of 1000−2000 Ω·cm , N-type Si with electric resistivity of 50−100Ω·cm, and P-type Si with electric resistivity of 50−100 Ω·cm. These samples with the three substrates were marked A, B, and C, respectively.

Interference microscope (Olympus BX51) and AFM (Chinese Academy of Sciences Chemistry Division CSPM3100) were used to analyze the macro- and micro-surface appearance. High-resolution XRD (Bede D1 system) was used to analyze the crystal characteristics. The X-ray source of the equipment is Cu Kα1 (λ=1.54056Å). The single crystal Si (220) surface is served as the reference crystal and analysis crystal. Based on different measurement requirements, the multiple crystals powder diffraction figuration model and the single crystal high-strength or high-resolution diffraction figuration model were adopted. Photoluminescence (PL) was used to study the luminescent properties of the sample, with a 410 nm semiconductor solid laser as the excitation source.

Results and Discussion

Interference microscope images of the samples are shown in Fig. 1. There are no cracks or circular defects resulting from the diffusion of the Si to the epitaxial layer in the three samples [16]; this appearance is observed in the whole epitaxial layer. This finding indicates that the lattice mismatch stress and thermal mismatch stress between the substrate and the epitaxial layer are relaxed without the appearance of macro defects. In the present paper, AlN is introduced as an interlayer, and a rich Ga

GaN buffer layer with low V/ III ratio was grown after producing AlN. Many studies show that a GaN epitaxial layer with better quality can be obtained after introducing an AlN layer. The introduction of AlN and a rich GaN double buffer layer with low V/ III ratio is the main reason that a GaN MQW LED epitaxial film with high performance was obtained in the present study. Crystal quality deviation dosage GaN is less than that of the GaN epilayer layer. However, as a buffer layer, it can play the role of relaxing the lattice mismatch stress and thermal stress between the substrate and the epitaxial layer, and laying the foundation for the growth of a high-quality epitaxial layer [17]. The entire surface of the three samples is neat without any large waves and grains. The surface of the A sample is rougher than that of the B and C samples, with small grains and waves. The sample surface on the P-type substrate is neater and has almost no waves and grains because the macro appearance of the epitaxial layer has a tendency to become refined.

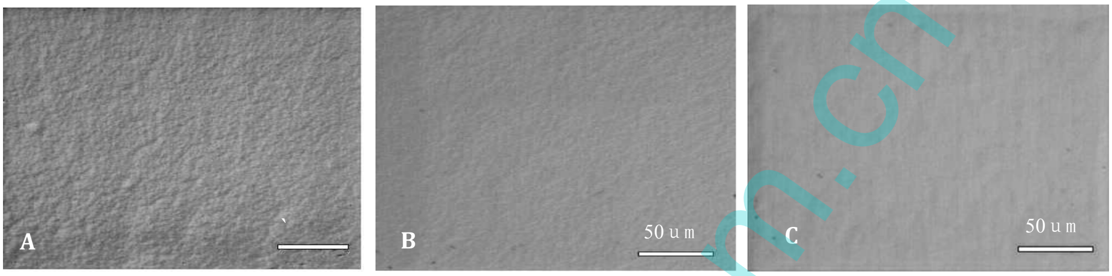

Fig. 1: Interference microscopic images of the samples grown on different substrates: Sample A; b) Sample B; c) Sample C

The AFM images of the samples are shown in Fig. 2. The roughness of the three samples is less than 3 nm: A is 2.9, B is 2.0, and C 2.5 is nm. Comparing A and B, the former has more waves on the surface and is rougher. B has larger hexagonal crystalline grains, a neat and smooth surface, and the least roughness. However, samples grown on the P-type substrate presents an entirely different N-type layered stack appearance, which could be caused by the different adulterants in different conduction substrates, resulting in the change in growing models of the GaN epitaxial film.

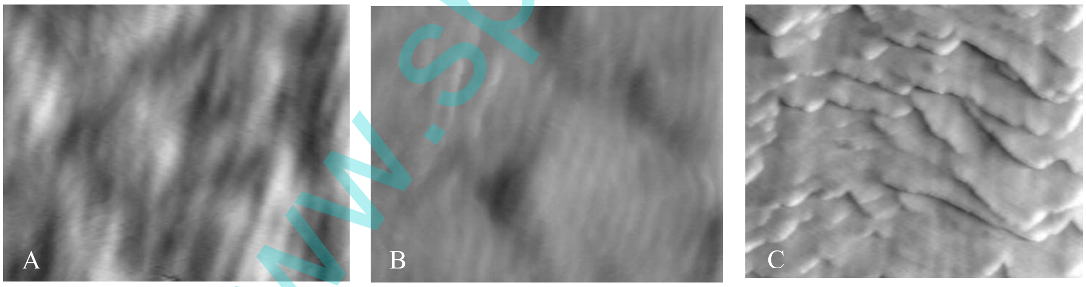

Fig. 2: AFM images of the samples grown on different substrates. Image range: 10×10 u m2. Roughness (RMS): a) Sample A: 2.9 nm; b) Sample B: 2.0 nm; c) Sample C: 2.5 nm

The DCXRD patterns are shown in Fig. 3. The symmetrical (002) surface and symmetrical tilt (102) surface of the samples were tested, respectively. The full width at half maximum (FWHM) of every diffraction peak is listed in Table 1. The FWHM of the Omega rocking curve of (002) and (102) surfaces of samples A and B is A: 361 and 496 arc s, and B:360 and 499 arc s, respectively, showing little difference. This result is comparable to the XRD result of GaN on the sapphire substrate, indicating that the GaN MQW LED epitaxial layer grown on the N-type Si substrate has better crystal quality. The FWHM of the Omega rocking curve of (002) and (102) planes of sample C is 375 and 540 arc s, respectively, which is obviously larger than that of samples A and B. This finding shows that the GaN crystal quality of the film on the P-type substrate is worse in comparing the films on the N-type substrate. Comparing the (002) plane FWHM value under the Omega scanning model with the Omega/2Theta scanning model, the former is greater than the latter; for (102), the value of the latter is greater than that of the former, depending on the stress state of the film. The FWHM of the Omega rocking curve mainly denotes the orientation information of the crystal, whereas the FWHM of the

Omega/2Theta rocking curve mainly reflects the heterogeneous strain and grain size. The rocking curve of the (002) surface corresponds to the C-directional information of the film, but the FWHM value of the Omega/2Theta (002) is smaller, indicating that the C-axial direction of the GaN received better relaxation. The rocking curve of the (102) surface combines the information from both A and C directions. The FWHM value of the Omega/2Theta (102) is larger than that of the Omega (102) because there is greater stress in the direction and heterogeneous strain under the action of a serious mismatch.

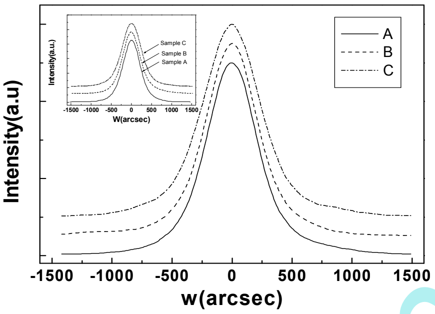

Fig. 3: X-ray Omega rocking curve of the GaN (0002) surface and GaN $(10\bar{1}2)$ surface. Illustration shows the corresponding Omega/2Theta rocking curve.

Table 1: Surface roughness and peak half-width of the DCXRD for each sample

Sample	RMS (nm)	FWHM (arcsec) (002 ω)	FWHM (arcsec) (002 ω-2θ)	FWHM (arcsec) (102 ω)	FWHM (arcsec) (102 ω-2θ)
A	2.9	361	247	496	514
B	2.0	360	241	499	523
C	2.5	375	252	540	560

The PL in room temperature of the samples is shown in Fig. 4. A blue light peak appeared in the field around 460 nm for each sample, but no peak appeared in other fields. This finding indicates that there is less light from the epitaxial layer defects. The PL peak values of the three samples are in the blue field, ranging from 460−470 nm. The PL peak wavelength of the A sample is 459 nm, with a peak half-width of 33.3 nm; those of the B sample are 460 and 33.8 nm, respectively; and those of the C sample are 467 and 37.1 nm, respectively. Comparing the peak wavelength of the A and B samples, that of the C sample has an red-shift caused by an insufficient relaxation of the C sample stress. Under stress, the blue-shift occurs in the peak wavelength of the PL. According to some research, the peak FWHM of the PL in room temperature of the InGaN/GaN MQW blue LED epitaxial layer on the Si (111) substrate is 35 nm [10]. In the present paper, the peak half-width values of the A and B samples are less than the reported values. The PL peak FWHM of the C sample is greater than that of the A and B samples due to the poor crystal quality of the C sample.

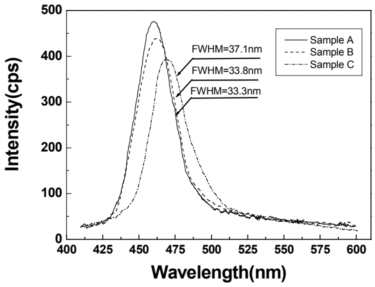

Fig. 4: PL spectrum of the samples at room temperature

Summery

With the introduction of AlN as an interlayer and the rich gallium GaN buffer layer with low V/III ratio, a InGaN MQW blue LED epitaxial layer without crack is grown on Si (111) substrate with three kinds of electric resistivity. The sample on the P-type substrate presents an entirely different layered stack compared with the sample on the N-type substrate with poor crystal quality. The peak positions of the PL spectrum are in the range of 460−470 nm, but the P-type substrate's wavelength and peak half-width are much more under greater stress. This finding indicates that the N-type Si (111) substrate is more suitable to the epitaxial GaN blue LED film than the P-type Si (111) substrate.

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