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The influence of AlN/GaN superlattice intermediate layer on the properties of GaN grown on Si(111) substrates*

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AlN/GaN superlattice buffer is inserted between GaN epitaxial layer and Si substrate before epitaxial growth of GaN layer. High-quality and crack-free GaN epitaxial layers can be obtained by inserting AlN/GaN superlattice buffer layer. The influence of AlN/GaN superlattice buffer layer on the properties of GaN films are investigated in this paper. One of the important roles of the superlattice is to release tensile strain between Si substrate and epilayer. Raman spectra show a substantial decrease of in-plane tensile strain in GaN layers by using AlN/GaN superlattice buffer layer. Moreover, TEM cross-sectional images show that the densities of both screw and edge dislocations are significantly reduced. The GaN films grown on Si with the superlattice buffer also have better surface morphology and optical properties.

Keywords: GaN, Si substrate, metalorganic chemical vapour deposition, superlattice buffer
PACC: 8115, 8115H

1. Introduction

Gallium nitride and related alloys are providing enormous benefits in optoelectronic- and electronic-devices.[1,2] GaN grown on silicon substrate has attracted great interest because Si is a low cost material and can be obtained easily in large size compared with other popular substrates, such as sapphire[3] and SiC.[4] Other advantage of using Si substrate is to offer a possibility to integrate GaN-based opto-electronics with Si-based micro-electronics. The GaN-based devices, like the light emitting diodes (LEDs)[5,6] and high electron mobility transistors (HEMTs),[7,8] have been fabricated on Si substrate. However, because of the high mismatch in lattice constant and thermal expansion coefficients between Si and GaN, the problems of cracks and high density of dislocations[9,10] still exist, which impede the fabrication of devices. The performance of the devices depends on the density of defects in the material. For example, threshold current, quantum efficiency and lifetime of nitride lasers have been improved by reducing the dislocation density. Moreover, melt-back etching will take place when Ga source contacts with Si at high temperatures.[11] The melt-back etching will heavily destroy the GaN surface morphology.

In previous reports, intermediate layers such as AlN,[12,13] AlGaN layer,[14,15] SiC layer,[16] $Al_xGa_{1-x}N$/GaN superlattice[17,18] and some new methods such as δ-doping,[19] ion-implant substrate,[20] and insertion of Si_xN_y layers were used to release tensile stress caused by large mismatch between Si substrate and GaN film. We have reported the accomplishment of crack-free GaN grown on Si(111) substrate by the use of AlGaN/AlN double buffers.[15] Here we use the superlattice as the buffer layer to grow high-quality GaN film on Si substrate. Insertion of a superlattice buffer layer is another effective and controllable method for improving the GaN quality. The role of superlattice buffer is to provide a compressive stress for GaN layer and compensate a part of tensile stress in the GaN epilayers. It is worth having a deeper understanding of the mechanism of stress release. In this paper, the interrelation between epilayers and buffers will be discussed in detail. The properties of GaN film with and without the AlN/GaN superlattice buffer layer have been compared, includ-

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ing the growth morphology, melt-back etching, stress condition, defects and optical properties. All these results will give a better understanding of the role of the superlattice layer in the growth of GaN on Si substrate.

2. Experiments

GaN films were deposited on p-type Si(111) substrates with about 5 cm in diameter. The growth was performed by using a vertical metalorganic chemical vapour deposition (MOCVD) system with Trimethylgallium (TMGa, flow of 40sccm), trimethylaluminium (TMAl, flow of 19 sccm), and ammonia (NH_3, flow of 6slm) as precursors. The substrates were cleaned by a wet chemical cleaning procedure, including boiling in H_2SO_4, H_2O:H_2O_2:NH_4OH=5:1:1 and H_2O:H_2O_2:HCl=4:1:1 successively to remove metal ions, grease, organic contamination, and then etched with HF(15%) for 1 min to remove the surface oxide layer. The growth process started with deoxidation of substrate surface under a H_2 flow at a high temperature of 1100°C. Two kinds of samples were grown, as shown in Fig.1. The first kind of sample only had a single AlN layer as buffer. The second kind of sample was additionally grown with the insertion of three sets of ten-period AlN/GaN superlattice buffer separated by 100nm GaN layers. The superlattice periods of AlN and GaN were 3 and 6 nm respectively. Choice of AlN and GaN layer thickness in the superlattice was made according to the critical thickness of the AlN/GaN system [21] and strain theory [22] to reduce the tensile stress of the GaN/Si system. For buffer layers and GaN epilayer the growth temperatures were selected to be 1000°C and 1050°C respectively. The reactor pressures were 1.33×10^4 and 2.66×10^4 Pa for buffer layers and GaN epitaxial layer respectively. In all samples the thickness of epitaxial GaN film was 1.2 μm.

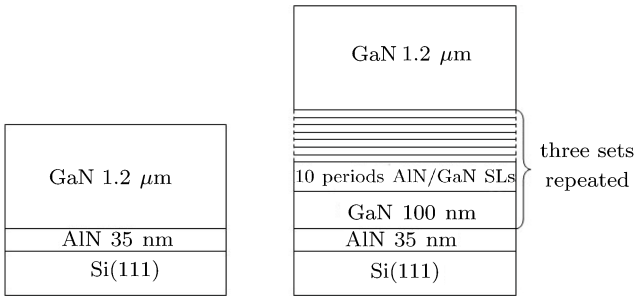


Fig.1. Schematic illustration of the two kinds of grown structures. (a) without AlN/GaN superlattice as insertion layers. (b) with AlN/GaN superlattice.

The GaN epilayers were characterized by atomic force microscope (AFM), optical microscope, Raman spectroscopy, transmission electron microscopy (TEM), and photoluminescence (PL). Surface morphologies of GaN films were recorded by AFM (Nanoscope IV) and optical microscope (Olympus) with CCD image. The TEM analysis of thinned sample was carried out in a JEM2010 TEM operating at 200kV. Room temperature micro-Raman scattering (JY-T64000) was used to investigate the stress status of the epitaxial layers in detail. For the micro-Raman scattering measurements, 488nm line of Ar-ion laser was employed as an excitation source, and backscattering polarization configuration was used. PL spectra were excited with the 325nm line of a He–Cd laser at the excitation power densities of 0.1–0.25 kW/cm^2.

3. Results and discussion

The surface morphologies of the 1.2 μm GaN films for two samples are given by AFM image, as shown in Fig.2. Apparently, the surface of the GaN films without superlattice buffer layer has a three-dimensional (3D) morphology. Some large voids can be observed in Fig.2(a). It is considered that these large voids are generated by the release of stress. While, with insertion of a superlattice buffer much better surface morphology is obtained. The size and density of the voids are greatly reduced. The roughness mean square (RMS) evaluated by AFM is remarkably improved from 2 nm to 0.3 nm on the 5×5 μm^2 area by introducing AlN/GaN superlattice buffer. Surface atomic steps can be clearly observed in Fig.2(b), indicating that a two-dimensional (2D) growth mode is obtained. The pinned defects are terminated at the surface steps, which are associated with the open core dislocations with the screw-type components.[22] This measurement is achieved by counting dark points, and we have the dislocation density on the surface of the GaN film with a superlattice buffer recorded by AFM is about 1.4×10^9 cm^{-2}, which is a fairly low value.

Cracks in GaN films have been investigated by optical microscope. Figure 3 shows the surfaces of GaN with and without the superlattice buffer layer. Although cracks cannot be avoided completely in both samples, the density of cracks in the GaN film with three sets of superlattices is significantly reduced (white bars in Figs.3(a) and 3(b) represent 20 and 200 μm respectively) and the crack-free area is about 30 mm^2.

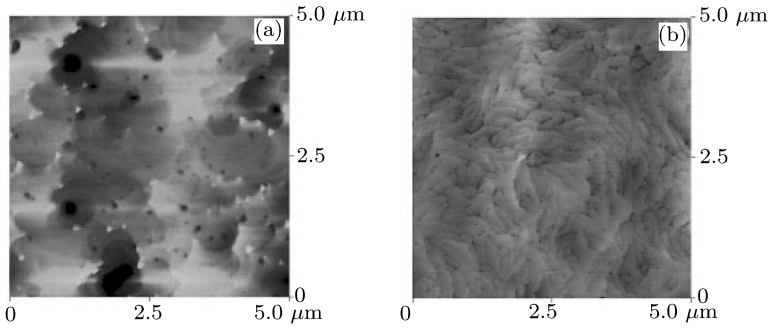


Fig.2. AFM images of the GaN/Si films on 5×5 μm^2 scanning areas. (a) without AlN/GaN superlattice. (b) with AlN/GaN superlattice.

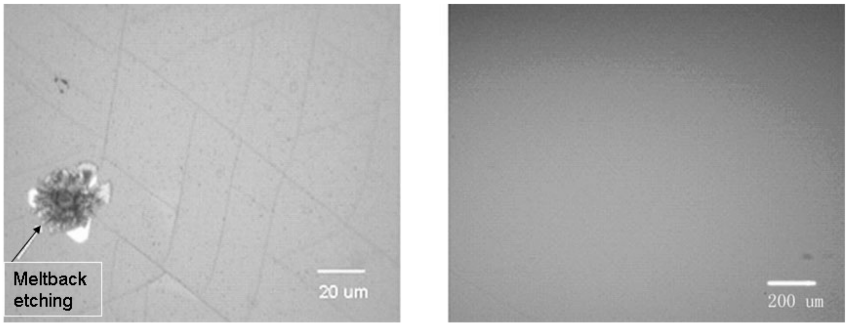


Fig.3. The optical microscope images of the GaN films on the areas that have most concentrated cracks density. (a) for the sample without superlattice buffer, (b) for the sample with AlN/GaN superlattice buffer (the white bar indicates 20 and 200 μm respectively).

The melt-back etching is another morphological defect in the GaN/Si material. In the case without superlattice buffer layer, the melt-back etching easily occurs during growth, as shown in Fig.3(a) indicated by an arrow. According to our experimental results, once the melt-back etching happens, the surface morphology of GaN films will be degraded with the growth going on. Fortunately, insertion of a superlattice buffer layer can eliminate the melt-back etching of GaN/Si. Because multi-set superlattice interlayer can effectively prevent the formation of large voids and deep cracks which are the main path for melt-back etching. In all the experiments we conducted, melt-back etching never happened on the GaN film having superlattice interlayer between GaN film and the Si substrate.

Through the above analysis, we see that GaN films have good surface morphology with the insertion of the superlattice buffer. Surface morphology has a very close relationship with stress conditions of GaN films. Raman spectra have been used to measure the phonon modes of GaN films and evaluate the stress in them (as shown in Fig.4).

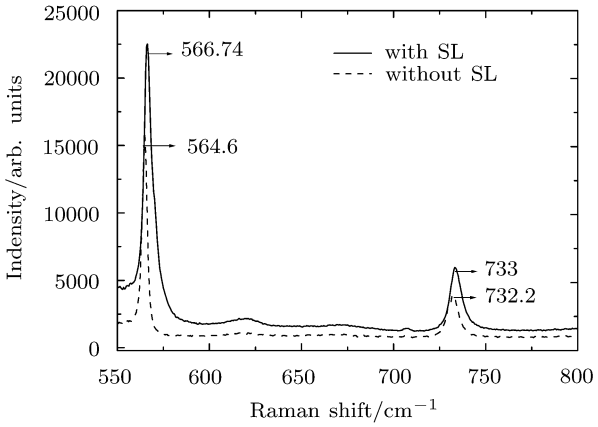


Fig.4. Raman spectra of the GaN films with and without AlN/GaN superlattice buffer.

The E_2 (high) mode red shift with respect to the standard value of 568 cm^{-1} implies a biaxial tensile stress in the epilayer. Here a stress coefficient k of 4.3 cm^{-1}/GPa is adopted.[23] The values of E_2 (high) for samples without and with the superlattice are 564.6 and 566.74 cm^{-1}, and the corresponding calculated

stresses are 0.79 and 0.29GPa, respectively; and the calculated stress value of 0.29 GPa is lower than the ultimate strength of breaking value 0.4 GPa under tensile stress for GaN.[24] This result shows that the tensile stress of GaN film is greatly released by inserting AlN/GaN superlattice buffer, which changes the stress conditions in the GaN film. Our results also are consistent with the conclusions obtained by using optical microscope.

The growth morphology is another very important material parameter. It can give the information of materials properties, such as interface, microstructure and growth defects. Here, TEM has been employed to characterize the growth morphology of GaN/Si. Figures 5(a)–5(d) show the cross-sectional bright-field images of the GaN grown on Si substrate, taken the $\langle 11\bar{2}0\rangle$ projection with diffraction vectors $\boldsymbol{g} = 0002$ and $\boldsymbol{g} = 1\bar{1}00$. According to the invisibility criterion of $\boldsymbol{g} \cdot \boldsymbol{b} = 0$ ($\boldsymbol{b}$ is Burgers vector), the image with $\boldsymbol{g} = 0002$ indicates that the threading dislocation (TD) has **c** component (screw dislocation), while the image with $\boldsymbol{g} = 1\bar{1}00$ shows that the dislocation has **a** component (edge dislocation). So the edge and screw components of dislocations can be visible. For GaN grown by MOCVD, the type **a** dislocation is predominant.[25] This type of TD originates from the climb and glide of basal-plane dislocations that are formed by Shockley partials in the faulted regions in the early buffer layer and can glide with the Burgers vector $\boldsymbol{b} = 1/3\langle 11\bar{2}0\rangle$.[26] Figures 5(a) and 5(b) show the screw and edge components of dislocations with a high density of $\sim 10^{10}\mathrm{cm}^{-2}$ in the GaN epitaxial layer of the sample without AlN/GaN superlattice buffer. A quite different effect can be observed in the sample with superlattice (in Figs.5(c) and 5(d)) that the TD density in the GaN epitaxial layer decreases substantially for both types of dislocations, especially for edge dislocations. From Figs.5(c) and 5(d), TD density is significantly reduced through the superlattice compared to early stages of growth in which the TD density is rather high. It can be explained by the fact that under the in-plane strain of the AlN/GaN superlattice, TD will move along the $1/3\langle 11\bar{2}0\rangle$ direction in the (0001) basal plane to create misfit dislocation, and is guided by the superlattice to the edge of the film. These results demonstrate clearly the role of the superlattice buffer in blocking TD propagation. Additionally, the enlarged image for superlattice areas exhibits good growth morphology of AlN/GaN superlattice with abrupt interfaces, as shown in the inset of Fig.5(d).

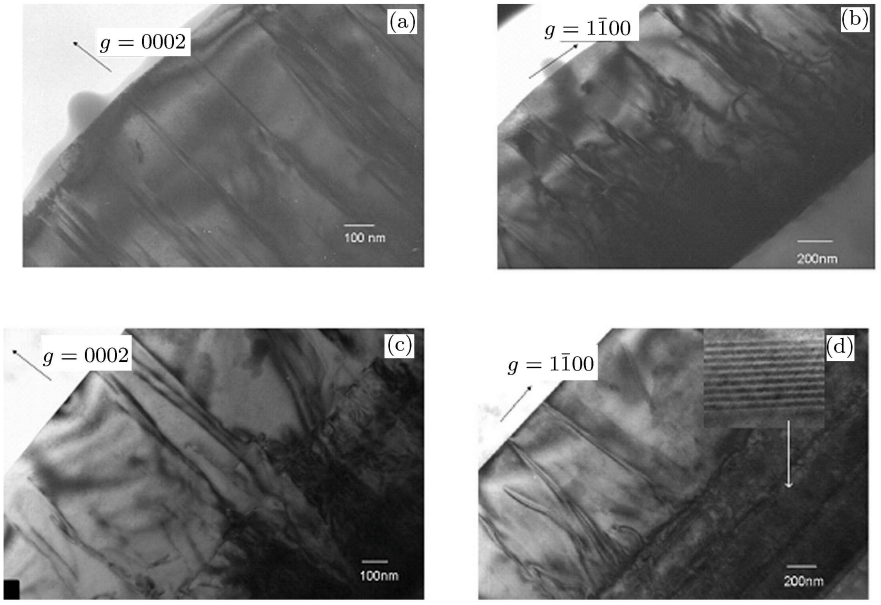


Fig.5. Fig.5 bright-field TEM image of the GaN/Si taken from a $\langle 11\bar{2}0\rangle$ projection. (a) without superlattice, $g = 0002$. (b) Without superlattice, $g = 1\bar{1}00$. (c) With superlattice, $g = 0002$. (d) With superlattice, $g = 1\bar{1}00$ (the inset in Fig.4(d) is the enlarged areas of the AlN/GaN superlattice).

The optical properties of the GaN samples have been characterized by PL measurements at room temperature. He–Cd laser is used as an exciting source. Band edge emission peak is observed at 3.35eV in both samples. This value shows the existence of red shift compared to that of GaN on sapphire, which is due to the lattice deformation resulting from the mismatch between GaN and Si substrate. The FWHMs of the band edge emission peak are 35 and 76 meV for GaN films with and without superlattice respectively. This result indicates that the optical properties of the GaN films with the use of superlattice buffer are improved, which benefits to the applications of LEDs or other optoelectronic devices.

4. Conclusions

In summary, the growth and characterization of GaN films grown by MOCVD on Si(111) substrates with and without superlattice have been studied. Crack-free GaN films have been obtained by insertion of superlattice layers. Raman shifts show a substantial decrease of in-plane tensile strain in GaN layers by using AlN/GaN superlattice buffers. Good surface morphology with 2D terraces and low RMS value of 0.3nm is observed. Both screw and edge types of dislocation densities are reduced significantly with the use of superlattice buffer. The results show that using AlN/GaN superlattice as buffer can greatly improve the optical properties of GaN epilayer grown on Si substrate.

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