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High-Quality and Strain-Relaxation GaN Epilayer Grown on SiC Substrates Using AlN Buffer and AlGaN Interlayer *

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We study the effect of the AlGaN interlayer on structural quality and strain engineering of the GaN films grown on SiC substrates with an AlN buffer layer. Improved structural quality and tensile stress releasing are realized in unintentionally doped GaN thin films grown on 6H–SiC substrates by metal organic chemical vapor deposition. Using the optimized AlGaN interlayer, we find that the full width at half maximum of x-ray diffraction peaks for GaN decreases dramatically, indicating an improved crystalline quality. Meanwhile, it is revealed that the biaxial tensile stress in the GaN film is significantly reduced from the Raman results. Photoluminescence spectra exhibit a shift of the peak position of the near-band-edge emission, as well as the integrated intensity ratio variation of the near-band-edge emission to the yellow luminescence band. Thus by optimizing the AlGaN interlayer, we could acquire the high-quality and strain-relaxation GaN epilayer with large thickness on SiC substrates.

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GaN and related alloys have attracted much attention for many years. Recent advances in GaN-based semiconductor thin film technology have paved the way for high-power ultraviolet to visible light-emitting diodes, laser diodes, ultraviolet detectors, and field effect transistors.[1,2] GaN has a band gap of 3.4 eV and forms continuous solid solutions with both AlN (6.2 eV) and InN (1.9 eV). Materials with engineered band gaps are envisioned for optoelectronic devices tunable in wavelength from the visible to the deep UV.[3] On the other hand, the relatively strong atomic bonding of these materials also points to their potential applications in high-power and high-temperature microelectronic devices. The numerous potential semiconductor applications of the wide band-gap Ⅲ–V nitrides have prompted significant research regarding their growth and development.

However, GaN-based semiconductors have a major drawback in terms of the lack of native substrates in large size and quantity.[1] Single-crystal wafers of GaN are not commercially available. Due to the lack of cheap and high-quality bulk GaN substrates, GaN epitaxial layers are still heteroepitaxially grown on sapphire (Al_2O_3), silicon carbide (SiC) or silicon substrates.[4] Sapphire is the most commonly used substrate, although its lattice parameter and coefficient of thermal expansion are significantly different from GaN. Si substrates offer extremely low cost and very large area wafer. However, it has been considered that the growth of high-quality GaN films on Si substrates has serious problems because of the cracks and high dislocation density of GaN epilayers due to the large mismatches in lattice constants and thermal expansion coefficient between GaN and Si substrate.[5–8]

To overcome negative lattice mismatch effects, silicon carbide has been used as a potential substrate. The similarity of the lattice structure of GaN and SiC, including closer lattice match and thermal expansion characteristics along the basal planes, is expected to improve structural quality of GaN materials.[9,10] However, we cannot directly grow GaN epilayers on SiC substrates due to the thermal mismatch between GaN and SiC, and cracks will be induced when a critical thickness of GaN is exceeded during cooling down.[11] In addition, micropipes in SiC substrates, as shown in Fig. 1, will decrease the crystal quality of the GaN epilayer. AlN is an ideal buffer layer for GaN epitaxy on SiC,[1,9] since it contains familiar structural characteristics and has small lattice mismatch with SiC. High temperature (HT) AlN is also considered as a relaxation layer to avoid cracks.[11] To modulate the strain during GaN growth on SiC, we adopt the AlGaN interlayer as a strain-modulation layer.

In this study, we report a 2 µm high-quality and crack-free GaN layer grown on (0001) 6H–SiC with AlN buffer and AlGaN interlayers, and we talk about the strain relaxation mechanism by AlN and AlGaN buffer layers.

The material used in this work was grown by metal organic chemical vapor deposition (MOCVD) on (0001) 6H–SiC substrates. Trimethylgallium (TMGa) and trimethylaluminum (TMAl) were used as precur-

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sors of Ga and Al, and ammonia (NH_3) was used as the nitride source. The substrate was first ramped to 1100℃ for hydrogen (H_2) baking to remove the surface damage which was induced by mechanical polishing.

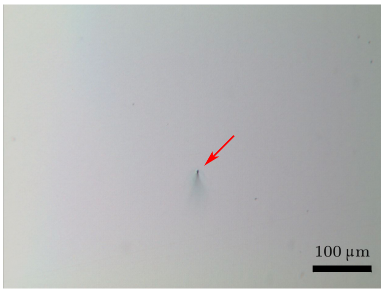

Fig. 1. The micropipe in the SiC substrate observed by an optical microscope with the magnification of 200X.

Samples A–D were prepared. The four samples were first deposited 100 nm AlN buffer at 1080℃, 100 Torr with 17.3 μmol/min TMAl, and 134 mmol/min NH_3. For samples B–D, an AlGaN interlayer with Al content 12% was introduced after AlN epitaxy. The AlGaN interlayer growth was at 1030℃, 100 Torr with 8.7 μmol/min TMAl, 34.4 μmol/min TMGa and 179 mmol/min NH_3. The thickness of the AlGaN interlayer for samples A–D were 0 nm, 5 nm, 10 nm and 20 nm, respectively. Finally, a nominally 2000-nm-thick GaN was deposited at 1030℃, 200 Torr with 258 μmol/min TMGa and 571 mmol/min NH_3 on four samples A–D. No intentional doping was introduced.

The crystalline quality of the GaN epilayer was estimated by high-resolution x-ray diffraction (XRD) rocking curves, which were obtained by using a Bede D1 XRD equipment. A 325 nm He-Cd laser was used as the excitation light source in the photoluminescence (PL) measurements, and a Horiba Jobin-Yvon Lab RAM HR800 Raman microscope was employed to measure the residual stress.

Symmetric (002) and asymmetric (102) XRD rocking curves were measured for examining the quality of GaN layers. Figures 2(a) and 2(b) are the (002) and (102) rocking curves for samples A–D. The full width at half maximum (FWHM) of the four samples as a function of the AlGaN interlayer thickness is shown in Fig. 2(c). Compared with sample A without the AlGaN interlayer, the FWHM of sample B with a 5 nm AlGaN interlayer slightly decreases for both (002) and (102), from 241 arcsec to 189 arcsec and from 279 arcsec to 240 arcsec, respectively, indicating a reduction of threading dislocations (TDs). As the AlGaN interlayer thickness increases to 10 nm for sample C, the FWHM of the (002) diffraction decreases to 129 arcsec slightly while the (102) diffraction decreases rapidly to 176 arcsec, indicating that the AlGaN interlayer can impede dislocations effectively. This value of GaN FWHM is better than the results reported.[12−14] When we further increase the AlGaN interlayer thickness to 20 nm, the FWHMs of (002) and (102) will not continue to decrease. From the results shown above, it is observed that the AlGaN interlayer could block dislocation effectively. However, when it exceeds a certain thickness, the dislocation density would increase again. We believe that it will lead to AlGaN layer relaxation when its thickness is too large, and this will introduce new dislocations for the GaN epilayer, thus the quality of GaN is not always improved with increasing the AlGaN interlayer thickness.

The FWHM of x-ray rocking curves has been used to quantify crystalline imperfection relatively. The rocking curves of symmetric planes are normally responsive to mosaic distortions but insensitive to the pure edge threading dislocation because these planes are undistorted by them.[15] Thus the (002) plane rocking curves are sensitive only to the screw and mixed-type threading dislocations. However, the rocking curves of the (102) asymmetry plane can be used to detect the crystalline distortions caused by all types of threading dislocations including pure edge dislocations.[16] The reductions of FWHMs in both directions suggest that all types of threading dislocations in the GaN film are reduced.

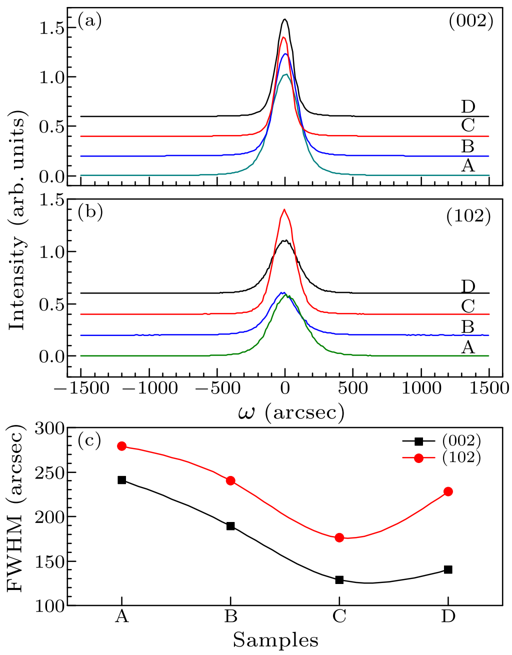

Fig. 2. XRD rocking curves of (a) (002) and (b) (102) of samples A–D. (c) The FWHMs of (002) and (102) of the four samples.

The surface of the GaN films exhibit different cracking situations for the four samples by an optical microscope, as shown in Fig. 3. For sample A, the GaN epilayer is almost cracked except the center area of the wafer. Cracks on sample B are less than sample A since an AlGaN interlayer was introduced in the former. Sample C has almost no cracks with the optimized AlGaN interlayer thickness. Sample D has almost the same situation of cracks as sample C. It is observed that cracks become less and less from sample A to sample D. We reckon that the AlGaN interlayer could adjust the stress and reduce the cracks for the GaN epilayer effectively.

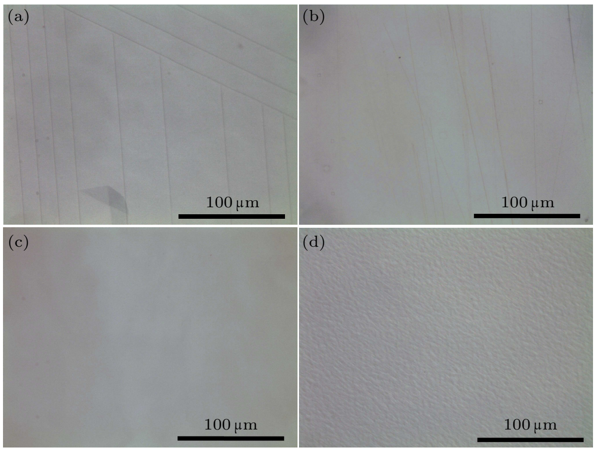

Fig. 3. Microscope images of samples A–D in (a)–(d), respectively.

Figure 4 represents the typical Raman spectra for the four samples. In the Raman scattering geometry, the GaN E_2 photon frequency has been extensively used to quantify the stress due to its sensitivity to strain. A tensile stress would generate a photon frequency red shift while a compressive stress must be responsible for a photon frequency blue shift. The strain could be valued by the formula of photon frequency shift of E_2 phonon mode in the light of the biaxial stress,[17]

$$\Delta\omega = k\sigma, \qquad (1)$$

where $\Delta\omega$ is the photon frequency shift (cm^{-1}), σ is the biaxial stress (GPa), and k is the Raman stress factor to be 2.7 cm^{-1}/GPa in this study.[18,19] Obviously, the biaxial stress can be calculated. The photon frequency shifts 1.54, 1.11, 0.83, 0.91 cm^{-1} are observed according to the phonon energy of E_2 mode at 568.0 cm^{-1} for a strain free bulk GaN[20] given a biaxial tensile strain of 0.570, 0.411, 0.307, 0.337 GPa for samples A–D, respectively. It is observed that the tensile stress in sample A is relatively larger than the other two samples, and this results in high crack densities in sample A.

The lattice mismatch between GaN and SiC is small and the lattice constant of GaN is slightly larger than that of SiC. Thus we attribute the large tensile stress presented in GaN to the effect of thermal expansion coefficient mismatch between GaN and SiC. Tensile stress was built up in the GaN film during sample cooling down from deposition temperature to room temperature after growth. This tensile stress could compensate for the compressive stress caused by lattice constant mismatch, thus the GaN epilayer would be cracked on the SiC substrate. The Raman results show that the biaxial tensile strain in the GaN layer is obviously released using an AlGaN interlayer. Combining with the GaN XRD results, we attribute the releasing of tensile stress in GaN to the threading dislocation (TD) reduction induced by the AlGaN interlayer. The compressive stress caused by the lattice mismatch between GaN and 6H–SiC in GaN could be partly relieved by generation of dislocations. From the XRD results, it is observed that the dislocations in GaN are obviously reduced by introducing the optimized AlGaN interlayer. As a result, the decrease of the dislocations would increase the residual compressive stress in GaN, which could offer enough compressive stress to compensate for the tensile stress caused by thermal mismatch during cooling down from growth temperature to room temperature.

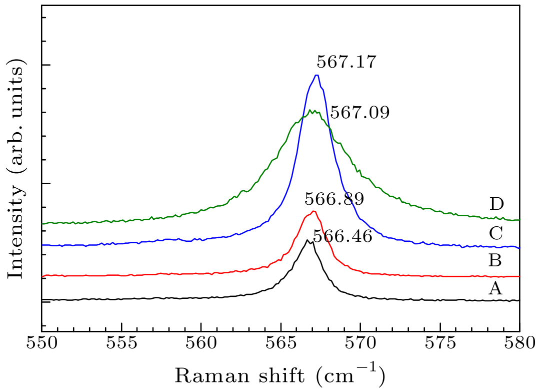

Fig. 4. Room-temperature Raman spectra near the E_2 mode of the four samples. A low-frequency shift indicates a tensile strain.

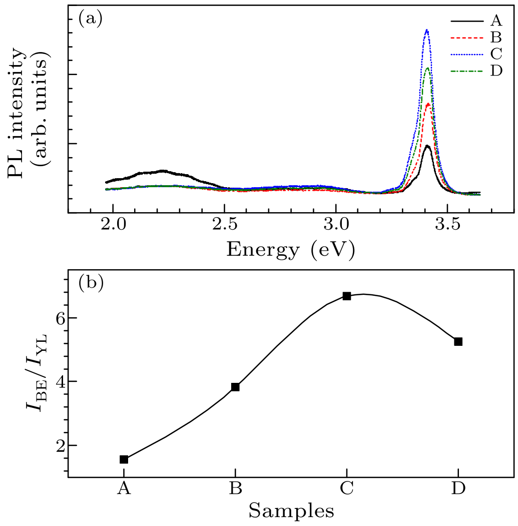

Fig. 5. (a) The room-temperature photoluminescence spectra of samples A–D. (b) The integrated intensity ratio of near-band-edge emission to the yellow luminescence band (I_{BE}/I_{YL}) of the four samples.

Figure 5(a) shows the room-temperature PL spectra of the four GaN samples. An obvious difference could be seen between the four PL spectra. In comparison with samples A, B and D, sample C with an optimized AlGaN interlayer thickness shows a stronger intensity of the near-band-edge emission, but a weaker yellow luminescence band. This means that the AlGaN interlayer and our optimized method have a remarkable influence on the optical properties of the GaN epilayer. The integrated intensity of near-band-edge emission and the yellow luminescence band (I_{BE}/I_{YL}) is shown in Fig. 5(b). The intensity of the near-band-edge luminescence increases rapidly from sample A to sample C and starts to decrease for sample D. The yellow luminescence band drops respectively, especially from sample A to sample B.

As shown in Fig. 5(b), the integrated intensity ratio of near-band-edge emission to the yellow luminescence band (I_{BE}/I_{YL}) increases from sample A to sample C. This suggests that there are less 'yellow luminescence defects' in the GaN samples from sample A to sample C. With the AlGaN interlayer thickness further increasing for sample D, the intensity ratio of I_{BE}/I_{YL} starts to drop. It is accepted that the threading dislocations may enhance the yellow luminescence in GaN.[21] Therefore, the changes in the PL intensities and the I_{BE}/I_{YL} ratio demonstrate a lower dislocation density for sample C and its structural quality may be significantly increased, which is consistent with the XRD testing results.

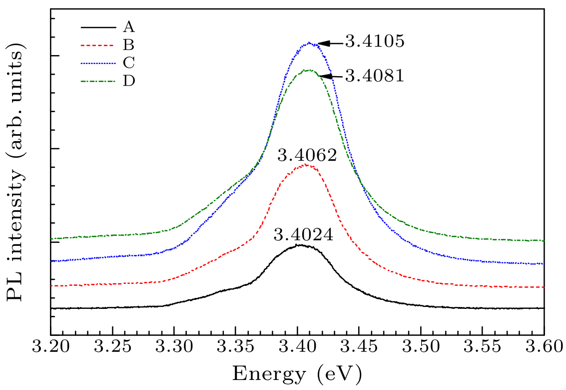

Fig. 6. The room-temperature photoluminescence spectra of the four samples.

It is known that the energy bandgap of GaN is affected by the residual stress in the film. A tensile stress would result in a decrease of the energy band gap while a compressive strain causes an increase of the band gap.[22] Figure 6 shows a set of PL spectra for the four samples. The bandgaps for samples A–D were 3.4024, 3.4062, 3.4105 and 3.4081 eV, respectively, while the bandgap for the free-standing GaN is 3.47 eV. Thus the three samples are in the tensile strain state. The variation tendency of the stress is the same as the measurements of the Raman scattering spectroscopy in Fig. 4.

In summary, the effect of an AlGaN interlayer on quality and strain of the GaN epitaxial films grown on SiC substrates has been studied. We obtain a GaN epitaxial layer over 2 µm which has high quality and less tensile stress. The XRD and PL (I_{BE}/I_{YL} ratio) measurements confirm that the dislocation density is decreased obviously with an optimized AlGaN interlayer. In addition to dislocation, the AlGaN interlayer is helpful to relieve tensile strain in GaN grown on SiC substrates, and the biaxial tensile stress is quantitatively analyzed by the Raman spectra, indicating that the tensile stress is reduced by an optimized AlGaN interlayer. Thus it is confirmed that the quality improvement and strain relaxation of the GaN epilayer grown on SiC are achieved by optimizing growth of the AlGaN interlayer.

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