Comparative study of different properties of GaN films grown on (0001) sapphire using high and low temperature AlN interlayers*

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Comparative study of high and low temperature AlN interlayers and their roles in the properties of GaN epilayers prepared by means of metal organic chemical vapour deposition on (0001) plane sapphire substrates is carried out by high resolution x-ray diffraction, photoluminescence and Raman spectroscopy. It is found that the crystalline quality of GaN epilayers is improved significantly by using the high temperature AlN interlayers, which prevent the threading dislocations from extending, especially for the edge type dislocation. The analysis results based on photoluminescence and Raman measurements demonstrate that there exists more compressive stress in GaN epilayers with high temperature AlN interlayers. The band edge emission energy increases from 3.423 eV to 3.438 eV and the frequency of the Raman shift of E_2(TO) moves from 571.3 cm^{-1} to 572.9 cm^{-1} when the temperature of AlN interlayers increases from 700 °C to 1050 °C. It is believed that the temperature of AlN interlayers effectively determines the size, the density and the coalescence rate of the islands, and the high temperature AlN interlayers provide large size and low density islands for GaN epilayer growth and the threading dislocations are bent and interactive easily. Due to the threading dislocation reduction in GaN epilayers with high temperature AlN interlayers, the approaches of strain relaxation reduce drastically, and thus the compressive stress in GaN epilayers with high temperature AlN interlayers is high compared with that in GaN epilayers with low temperature AlN interlayers.

Keywords: GaN, AlN interlayers, high temperature
PACC: 7280E, 7865K, 8115H

1. Introduction

III-nitride semiconductor materials are attractive and have been studied for many years because of its advantages such as the large breakdown voltage of wide gap materials, high electron mobility, high stability of temperature and chemistry.[1,2] GaN-based materials are used extensively in optoelectronics and high temperature, high frequency and high power electronics, such as AlGaN/GaN high electron mobility transistors (HEMTs). In the past decades, remarkable breakthroughs have been made in their growth and applications in blue laser diodes (LD), high-bightness light emitting diodes (LED), ultraviolet photo-detectors. However, there remain many problems regarding the growth of high crystalline quality GaN films, which have not been well resolved.

The lack of native GaN substrate has always hindered the growth of high quality GaN films. Most of the GaN layers are grown on (0001) sapphire substrate by metal organic chemical vapour deposition (MOCVD) in spite of some serious limitations, such as insulating property, the unavailability of large sizes and extreme hardness. Due to a large lattice mismatch (16%) and a big difference in thermal expansion coefficient between GaN and sapphire, it is more difficult to grow high quality GaN films on sapphire and there always appear 10^8–10^{10} cm^{-2} threading dislocations (TDs) in it. A very high TD density has a negative effect on the device performances and the property of GaN epilayers. For example, TD lines act as Coulomb scattering centres in GaN epitaxial films, and the transverse mobility at low carrier concentration is limited by scattering at the dislocation lines in AlGaN/GaN HEMTs.[3] It was also reported that the leakage current was related to the dislocation density in GaN-based LED.[4] Reduction of TDs is indispensable for improving crystalline quality and prolonging device lifetime and enhancing characteristic. In order to achieve high quality GaN films, various

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growth structures and methods have been developed, such as using a low temperature (LT) GaN interlayers (ILs),[5] LT AlN ILs,[6] porous AlN ILs[7] and using SiN mask,[8] epitaxial lateral overgrowth (ELO),[9] pendeo epitaxy (PE), selective area growth (SAG).[10] LT AlN IL is a very promising way to reduce TDs in GaN epilayers, and it is quite often used. However, high temperature AlN ILs has been seldom used and the effect of the growth temperature of AlN ILs on GaN epilayers is unknown.

In this paper, we report on the different properties of the GaN films with HT and LT AlN ILs grown on (0001) sapphire substrate by MOCVD technique. The material crystalline quality of the GaN epilayers is studied by using high resolution x-ray diffraction (HRXRD). Photoluminescence (PL) and micro-Raman spectroscopy are used to characterize the residual stress of GaN films.

2. Experimental details

The GaN epilayers grew on (0001) sapphire substrates in a home-made vertical-type MOCVD system. Trimethylauminum (TEA), trimethylgallium (TMG) and ammonia (NH_3) were used as precursors of Al, Ga, and N, respectively. Purified H_2 was used as the carrier gas. First, a low temperature AlN seed layers, about 25 nm thick, grew at 560 °C. Subsequently an approximately 500-nm-thick GaN layers grew at 1050 °C followed by a 20-nm-thick AlN ILs at different temperatures. After that, a 1-μm-thick GaN top layer grew on the AlN ILs. The temperatures of AlN ILs were 700 °C and 1050 °C for LT-AlN and HT-AlN ILs samples, respectively. The schematic diagrams of two GaN samples of AlN ILs at different temperatures are shown in Fig. 1.

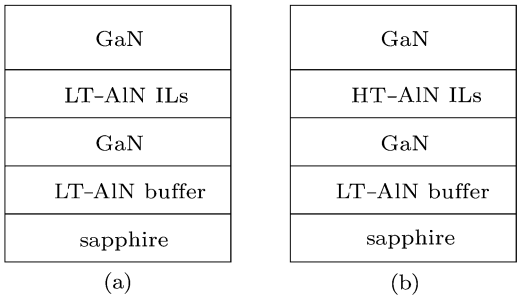


Fig. 1. Schematic diagrams of two GaN samples with (a) LT-AlN and (b)HT-AlN ILs.

After the growth, a series of characterization techniques, including high resolution x-ray diffraction (HRXRD), photoluminescence (PL) and Raman spectroscopy, were used to investigate the crystalline quality and optical properties of the samples. Measurements of HRXRD were performed by using monochromated Cu-Kα_1 radiation ($\lambda = 1.5405$ Å, 1 Å=0.1 nm) on a high resolution diffractmeter D8 discover system from Bruker-AXE, equipped with an asymmetric 2-bounce Ge (220) monochromator and a scintillation detector. PL was measured with a 25 mW He–Cd laser operating at 325 nm as an excitation source. Raman spectroscopy was performed by a Jobin Yvon microscopy with a 488 nm line of the argon ion laser with $Z(XX)-Z$ configurations geometry and its lateral spatial resolution is 1 μm. All measurements were performed at room temperature.

3. Results and discussion

In order to investigate the crystalline quality of GaN films, HRXRD was performed in samples by using an $\omega - 2\theta$ scan mode. Figure 2 shows typical x-ray rocking curves of (0002) and $(10\bar{1}2)$ diffractions of GaN films. The values of full width at half maximum (FWHM) of the x-ray rocking curve of (0002) for GaN epilayers with LT AlN ILs and HT AlN ILs are 478 arcsec and 383 arcsec, respectively, and the values of FWHM of $(10\bar{1}2)$ rocking curve are 928 arcsec and 707 arcsec. It appears that the crystalline quality of GaN layers increases significantly by the insertion of HT AlN ILs.

As is well known, there are three types of threading dislocations in GaN epilayers, i.e. screw type, edge type and mixed type. The title range of the lattice planes for wurtzite GaN films reflects the extent of screw dislocation with a Burgers vector $b =$ (0001), and the twist of lattice planes reflects the extent of edge-type dislocation with a Burgers vector $b = 1/3\,(11\bar{2}0)$. The FWHM of rocking curves can reflect the crystalline quality of GaN films, and we can estimate the dislocation density D of GaN films by the following equation:[11]

$$D = \frac{\beta^2}{9b^2},$$

where D is the dislocation density, β is the FWHM measured by XRD rocking curve, and b is the Burgers vector length ($b_{\rm screw} = 0.5185$ nm, $b_{\rm edge} = 0.3189$ nm). The screw dislocation density decreases

from 2.2×10^8 cm^{-2} to 1.4×10^8 cm^{-2} and edge dislocation from 2.2×10^9 cm^{-2} to 1.3×10^9 cm^{-2} as the temperature of AlN ILs increases from 700 °C to 1050 °C. From the results, we can conclude that the suppression and the interception of dislocation by HT AlN ILs are more effective, especially for the edge dislocation. To figure out the influence of temperature difference of AlN ILs on the crystalline property, we need to analyse the growth mechanism of GaN films on AlN ILs.

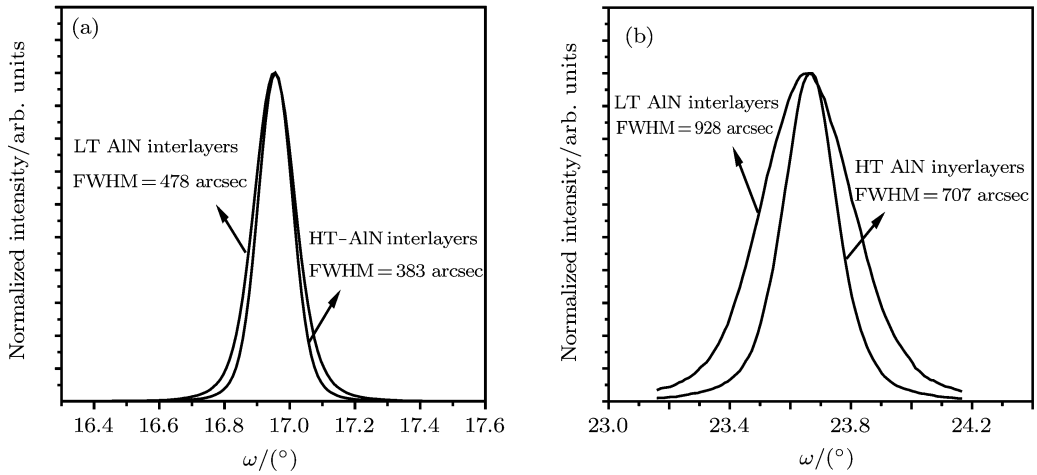


Fig. 2. Rocking curves of (a) (0002) and (b) ($10\bar{1}2$) diffractions of GaN epilayers with HT and LT AlN ILs.

Considering the fact that GaN films are formed by the coalescence of the crystalline grains during the GaN growth at high temperature, and the threading dislocations are mainly located in the zones of coalescence, we believe that the temperature of AlN ILs can account for the morphology and the distributions of threading dislocations in GaN layers. The AlN grains will provide islands for the growth of subsequent GaN at the initial stage. After growing at both lateral and vertical directions (three-dimensional growth mode), the islands will coalesce. The coalescence rate depends on the size and the density of the islands. When a smooth layer of GaN is established, the GaN begins to grow in a quaasi-2D mode.

When growing at a low temperature (700 °C), AlN always forms an amorphous structure. The island size is small, and the island density is high. The spaces between the islands are too small to grow in a lateral direction, all threading dislocations have to vertically extend to surface in the course of GaN film growth. However, because of high temperature of AlN ILs growth, the nucleation island size is large,[12] and the island density is low. The spaces between the islands are so large that almost all islands grow in the lateral direction. So the threading dislocations have sufficient spaces to be bent and interactive when extending to the surface. This may explain the reason why the crystalline quality of GaN epilayers with LT AlN ILs is worse than that with HT AlN ILs.

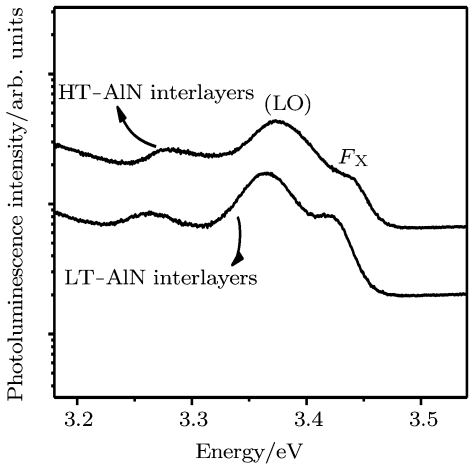


Fig. 3. Photoluminescence spectra of GaN samples with HT and LT AlN ILs.

Meanwhile, in order to estimate the residual stress in GaN films, PL and micro-Raman measurements were carried out on GaN samples. Figure 3 shows the room temperature PL spectra of GaN samples with HT and LT AlN ILs, showing obvious blue shift to-

wards to high energies. The band edge emission energy of GaN films corresponding to the recombination of free exciton F_X increases from 3.423 eV to 3.438 eV and the longitudinal optical (LO) phonon replica of F_X increases from 3.365 eV to 3.375 eV when the temperature of AlN ILs growth increases from 700 °C to 1050 °C. Due to the blue shift of PL spectrum corresponding to compressive strain, the GaN epilayers with HT AlN ILs contain higher compressive strain.[13]

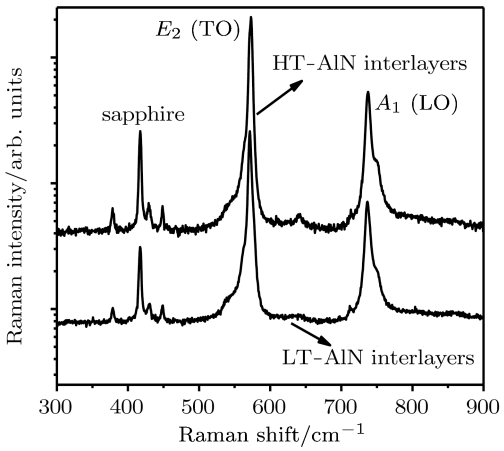


Fig. 4. Raman spectra of GaN epilayers with HT and LT AlN ILs .

Figure 4 shows the room temperature Raman spectra of GaN epilayers with HT and LT AlN ILs. Figure 5 exhibits the expanded spectra of E_2 (TO) peak area measured at room temperature. Hexagonal GaN has a wurtzite structure and belongs to the C_{6v}^4 space group. Group theory predicts eight sets of phonon modes: $2A_1 + 2E_1 + 2B_1 + 2E_2$ at the Γ points of the Brillouin zone. One set of A_1 and E_1 modes are acoustic. One A_1 mode, one E_1 mode, and two E_2 modes are Raman active. Two B_1 modes are silent. The A_1 and the E_1 symmetries are polar and split into longitudinal (LO) and transverse (TO) components. Then there are six first-order Raman-active optical modes at the Γ point: $A_1(\mathrm{TO})+A_1(\mathrm{LO})+E_1(\mathrm{TO})+E_1(\mathrm{LO})+2E_2$ in various scattering configurations. According to Raman selection rules, only E_2(TO) and A_1(LO) modes should be observed with the $Z(XX)-Z$ scattering geometry in our experiments.

In Raman spectrum, the peak at 418 cm^{-1} is caused by the phonon from the sapphire. Other two peaks can also be found, which are associated with the E_2(TO) and the A_1(LO) peaks of GaN films. The E_2(TO) phonon of GaN is sensitive to the change of strain state in it and widely used to assess the strain in c-plane parallel direction, which can be accurately measured by using Raman spectroscopy. As we can seen from Fig. 5, the Raman shift of E_2(TO) phonon of GaN epilayers with HT and LT AlN ILs are 572.9 cm^{-1} and 571.3 cm^{-1}, respectively. An increase in the E_2(TO) photon frequency with respect to unstrained GaN indicates compressive stress, whereas a decrease suggests the occurrence of a tensile stress. Both of GaN films have compressive stresses because the E_2(TO) phonon frequency shifts toward high frequency compared with the 567.5 cm^{-1} for a 400 μm free standing bulk GaN layer. The shifts of E_2(TO) phonon frequency of GaN epilayers with HT AlN ILs are larger than those with LT AlN ILs, which means that the GaN epilayers with HT AlN ILs are under more compressive strain in the parallel direction. The relation between the residual stress ε and the measured Raman shift of the E_2(TO) phonon line $\Delta\omega$ is derived to be[14]

$$\varepsilon = \frac{\Delta\omega}{k},$$

where ε is in-plane strain, and k is the linear stress-shift coefficient ($k = 4.2$ GPa^{-1}·cm^{-1}). So the values of compressive stress in GaN epilayer with HT and LT AlN ILs are 1.29 GPa and 0.9 GPa, respectively. The conclusions drawn from Raman spectrum are consistent with the above-mentioned PL measurements.

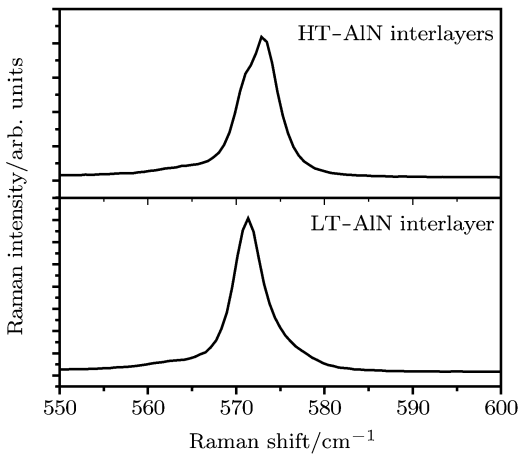


Fig. 5. Expanded spectra of E_2(TO) phonon peak area measured at room temperature.

To further determine the effect of the temperature of AlN IL on the property of GaN epilayer, we can explore the relationship between crystalline quality and strain state in GaN epilayers. These consequences might be due to the effect of strain from the thin inserted AlN ILs on the dislocation interactions in

the compressively strained GaN epilayers. As is well known, the most common mode of misfit stress relaxation is related to the formation of misfit dislocations at the layer/substrate interfaces. The contribution of the dislocation inclination to the stress relaxation is primary in mismatched layers. Due to the low quality of GaN epilayers with LT AlN ILs, the density of threading dislocation in it is high compared with those with HT AlN ILs, and the strain relaxation is sufficient. Hence the residual compressive stresses in GaN epilayers with HT AlN ILs are larger than those with LT AlN ILs.

In summary, a model for the formation of the threading dislocation morphology, with the growth mechanism of GaN films taken into consideration, is put forward to explain the variations of density of threading dislocations in GaN, especially, the relation between the growth mechanism and the variations of threading dislocations density with temperature of AlN IL varying. In our model, it is believed that the island size and the island density are determined by the growth temperature of AlN IL and can directly determine the growth directions of TDs in subsequent GaN growth.

4. Conclusions

Comparative study on high and low temperature AlN interlayers (ILs) and their roles in the properties of GaN epilayers growing on (0001) plane sapphire substrates by means of MOCVD are carried out. Based on the measurements by HRXRD, PL and Raman spectroscopy and analysis, it is concluded that crystalline quality is improved and the threading dislocations are reduced significantly by using HT AlN ILs compared with using LT AlN ILs, especially for the reduction of the edge type dislocations. Meanwhile, there exists a larger residual compressive stress in GaN epilayers with HT AlN ILs. It is concluded that the growth temperature of AlN ILs effectively controls the size and the density of the nucleation islands for subsequent GaN growth. For the GaN epilayers with HT AlN ILs, the nucleation islands have large sizes and low densities, which is an advantage to threading dislocation bent and interaction when it extends to the GaN epilayer surface. So the threading dislocation density is reduced and crystalline quality is improved. Due to the main stress relaxation resulting from the formation of TDs, the GaN epilayers with HT AlN ILs each contain a higher compressive stress with a low density of threading dislocations. Therefore, the growth method with using HT AlN ILs can improve crystalline quality and increase the compressive stress of GaN epilayers, which is beneficial to and desirable in the epitaxy of nitride devices structures used in HEMTs and can increase the density of two-dimensional electron gas.

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