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Growth studies of m-GaN layers on $LiAlO_2$ by MOCVD

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This paper reports that the m-plane GaN layer is grown on (200)-plane $LiAlO_2$ substrate by metal-organic chemical vapour deposition (MOCVD) method. Tetragonal-shaped crystallites appear at the smooth surface. Raman measurement illuminates the compressive stress in the layer which is released with increasing the layer's thickness. The high transmittance (80%), sharp band edge and excitonic absorption peak show that the GaN layer has good optical quality. The donor acceptor pair emission peak located at ∼3.41 eV with full-width at half maximum of 120 meV and no yellow peaks in the photoluminescence spectra partially show that no Li incorporated into GaN layer from the $LiAlO_2$ substrate.

Keywords: $LiAlO_2$ substrate, m-plane GaN layer, optical properties
PACC: 6855, 7280E, 0765G

1. Introduction

There is huge interest worldwide at present in finding an alternative substrate for GaN-based epitaxial deposition. Currently, silicon carbide (SiC) and sapphire are the favourite wafers for the AlGaInN material system. These substrates suffer from the limitations that growth is only possible along the polar c-axis which leads to strong piezoelectric and spontaneous polarization fields. The band bending related to polarization-induced fields is responsible for the poor electron–hole overlap. To achieve highly efficient quantum well structures, growth along non-polar axes is required. The m-plane growth direction demonstrates one of non-polar axes for the deposition of the nitrides, it prevents the band bending and provides direct recombination across the whole well.[1−4]

$LiAlO_2$ (LAO) substrates, with their huge advantage of low cost and easy substrate removal, offer the possibility to grow GaN along the m-plane direction. The growth of GaN on this substrate has been investigated by several research groups.[5−11] Recently two groups have demonstrated their m-plane blue light emitted diodes (LEDs) grown on LAO substrates.[12,13] Now, we also fabricate m-plane blue LEDs grown on LAO substrates. In this paper, the GaN layers were studied in detail by x-ray diffraction (XRD), AFM, Raman spectra, absorption spectra and photoluminescence (PL) spectra.

2. Experimental details

Specular GaN films were grown on (200)-plane LAO substrates in a vertical close-spaced, rotating disc MOCVD reactor. The LAO substrates were nominal (200)-plane wafer (±0.2° surface orientation tolerance) obtained from the crystal grown by us.[14−21] The as-received substrates were degreased sequentially in trichloroethylene (10 min), acetone (5 min), and methanol (5 min) in an ultrasonic bath, finally blown for dryness with filtered nitrogen. The growth process was modelled after the two-step process that has become the standard for the growth of c-GaN on c-sapphire. The LAO substrates were annealed *in situ* prior to the deposition of a low temperature GaN nucleation layer (about 0.1μm) at ∼850°C. The temperature was then raised to ∼1100°C and epitaxial GaN films approximately 0.6μm thick were grown at rates

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ranging from 2Å/s to 3Å/s. Trimethylgallium and ammonia were used as the group III and V sources respectively. Raman measurements are performed at room temperature by focusing the 632 nm line of an Ar^+ ion laser on the sample and detecting the backscattered light. The spectral resolution is about $1\,cm^{-1}$. XRD was performed on a Japanese D/max 2550V x-ray diffractometer to assess the crystallinity of the thick film, and an on-axis high-resolution XRD was carried out on a Philips four-circle x-ray diffractometer operating in receiving slit mode with four bounce Ge (220)-monochromated CuKα radiation and a 1.2 mm slit on the detector arm. The surface morphologies were observed with atomic force microscope (AFM) (Veeco, NanoIIIa multimode). The optical transmission was examined at room temperature with a JASCO V-570 UV/VIS/NIR spectrophotometer in the wavelength range from 190 to 1900nm. PL spectra were obtained by using He–Cd laser as a light source using an excitation wavelength of 325 nm at room temperature.

3. Discussion and results

Figure 1 shows the $\theta - 2\theta$ scan XRD pattern of GaN/ LAO (200). It was revealed that hexagonal GaN film with a strong preferred $[1\bar{1}00]$ orientation was grown on LAO (200). The sharp peak at 16.142° stems from GaN $(1\bar{1}00)$ reflection, demonstrating that the fraction of c-plane GaN, if any, is indeed very small, while the sharp peak of unstrained GaN locates at 16.19°. It was revealed that hexagonal GaN films with a strong preferred $[1\bar{1}00]$ orientation can be

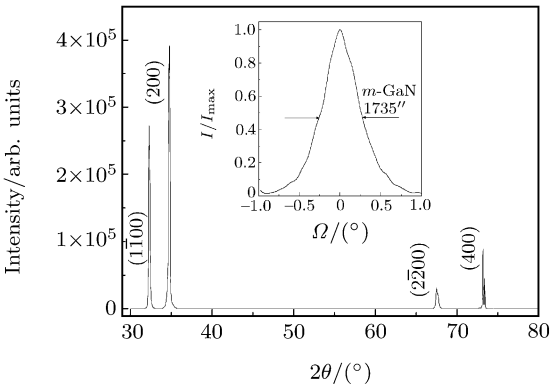


Fig.1. X-ray diffraction profile and its rocking curve of the $(1\bar{1}00)$ reflection for the 700nm thin GaN film grown on $LiAlO_2$ (200) substrate.

grown on LAO (200) substrate. The diffraction angle for GaN $(1\bar{1}00)$ evidences a dilation of 0.29% parallel to the growth direction $[1\bar{1}00]$. This means that compressive stress exists in m-GaN layer. XRD rocking curves of the m-GaN films exhibit a full-width at half maximum (FWHM) value of 1735″, shown in the inset of Fig.1. These results mean that high-quality $(1\bar{1}00)$ GaN can be grown on (200) LAO substrate by MOCVD method.

The surface morphology of non-polar GaN films observed by AFM is shown in Fig.2. The m-plane GaN surface appears tetragonal-shaped crystallite, the largest crystallites are about 1000nm×200nm in square and 80nm high. Therefore it shows a smooth surface with a root mean square roughness of 100nm. The morphology partially reflects that m-plane GaN has good crystallinity and little stress, this is in agreement with the results of x-ray rocking curve.

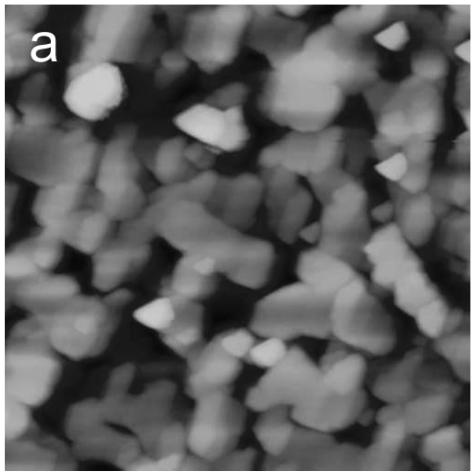


Fig.2. 5μm×5μm AFM images of surface features for m-plane GaN layer.

Figure 3 shows measurements of 0 and 0.6μm depth GaN film (the GaN surface and the buffer layer) in $y(x,x)\bar{y}$ scattering configuration, where x, y correspond to the $[11\bar{2}0]$ and $[1\bar{1}00]$ directions for m-plane GaN. Information on stress and the crystalline quality can be extracted from the E_2 phonon frequency and linewidth in Raman spectra. The width of the E_2 Raman peak reflects the crystalline quality. Stress affects the E_2 phonon frequency. Unstained GaN is characterized by E_2 frequency of $567\,cm^{-1}$.[22–24] An increase in the E_2 phonon frequency with respect to unstained GaN indicates compressive stress, whereas a decrease points to tensile stress. Compressive and tensile stress are commonly found in thin GaN films grown on sapphire, on SiC substrates, and so on. From our figures, it was found that the E_2 phonon frequencies shifted from the $575.15\,cm^{-1}$ to $574.59\,cm^{-1}$ for m-GaN film and the width became narrower when

the measured point shifted from buffer layer (0.6μm depth) to GaN film surface (0μm depth), which shows that the crystallinity of the layer gradually improves with increasing thickness because of lattice-mismatch or thermal mismatch relaxation between the substrate and the layer.

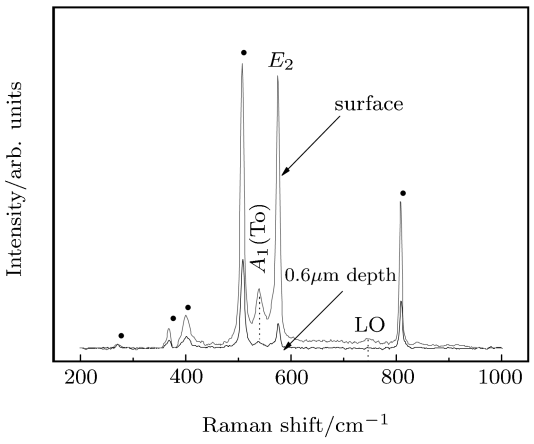


Fig.3. Micro-Raman spectra of m-GaN film on (200) $LiAlO_2$, an asterisk refers to peaks originating from the substrate.

The crystallinity of GaN film was also investigated by measuring the transmittance and the absorption. The transmittance of m-GaN film was 80 % in the wavelength range of 400–800 nm. The fall of transmittance was very sharp near absorption edge, which indicates high crystallinity. Besides, in absorption spectra at RT shown in Fig.4 excitonic absorptions are clearly visible for m-GaN films fabricated on (200) LAO. Excitonic absorption spectra are observed in the films even though the absorption spectra do not split into two or more peaks. This is also sufficient to display that the crystallinity of GaN films is so high that inhomogeneity-induced broadening effect obscuring the observation of excitonic peak at RT can be suppressed. The band gaps were obtained by plotting α^2 vs $h\upsilon$ from the absorption spectra using the relation $\alpha^2 \propto (h\upsilon - E_g)$, and extrapolating the straight line portion of this plot to the energy axis where α is the absorption coefficient and $h\upsilon$ is the photon energy. The optical band gaps of m film shown in the inset of Fig.4 were estimated to be 3.436 eV. For stoichiometric GaN films, the optical band gap is known to be about 3.4 eV. Therefore, the obtained optical band gaps demonstrate high crystallinity of the m-GaN film on LAO.

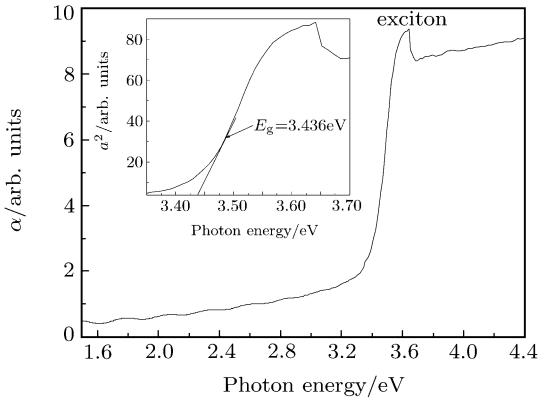


Fig.4. Absorption spectra measured at room temperature for m-GaN films and plot of α^2 as a function of $h\upsilon$ in the inset.

The optical properties of the GaN layer were also measured by PL at room-temperature as shown in Fig.5. We observed the strong band-edge emission at RT. The donor acceptor pair emission peak energy is ~3.41 eV with FWHM of 120 meV. The peak shows little red shift at band-edge emission peak. It seems that the GaN grown by MOCVD is under some compressive stress state, which is related to XRD and Raman spectra results. No yellow peaks at ~2.23 eV were observed. Kung *et al*[25] and Pankove *et al*[26] has reported the PL peak at ~2.23 eV at room temperature probably rooted in the incorporation of Li into GaN as a result of the decomposition of the substrate at elevated temperatures. The results show that no Li incorporated into GaN layer fabricated by MOCVD method. Our results are quite challenging to grow the high-quality GaN layer on (200) LAO substrate by MOCVD method for device application.

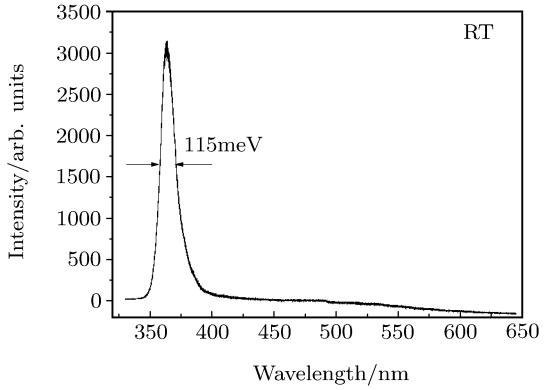


Fig.5. Photoluminescence spectra obtained at room temperature for m-GaN layer.

4. Conclusions

We have grown m-plane GaN layer on (200) $LiAlO_2$ substrate by MOCVD method. Tetragonal-shaped crystallites observed by AFM appears at the smooth surface. The little compressive stress in the layer was released and the crystal quality was greatly improved with increasing the layer's thickness. The optical properties were examined by absorption spectra and PL spectra. We find that the layer has high transmittance (∼80%), sharp band edge and observed excitonic absorption peaks. Moreover, it has only the donor acceptor pair emission peak located at ∼3.41 eV with FWHM of 120 meV. No yellow emission peak proves that no Li incorporated into GaN layer which partly shows that the $LiAlO_2$ was undecomposed during the GaN growth. Our results illuminate that high-quality m-plane LEDs can fabricate on LAO substrate by MOCVD method, which was reported in other paper.

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