Effect of growth temperature of AlN interlayers on the properties of GaN epilayers grown on c-plane sapphire by metal organic chemical vapor deposition

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Received 31 August 2009, accepted 2 March 2010
Published online 21 June 2010

Keywords AlN, GaN, MOCVD, growth, structure, dislocations, strain relaxation, Raman spectra

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The effect of growth temperature of AlN interlayers on the properties of GaN epilayers grown on c-plane sapphire by metal organic chemical vapor deposition has been investigated by high resolution X-ray diffraction (HRXRD) and Raman spectroscopy. It is concluded that the crystalline quality of GaN epilayers is improved significantly by using the high temperature AlN (HT-AlN) interlayer in GaN buffers. The density of threading dislocation is reduced especially for edge type dislocations. Higher compressive stress exists in GaN epilayers with HT-AlN interlayer than with low temperature AlN (LT-AlN) interlayer, which is related to the reduction of strain relaxation caused by the formation of misfit dislocation.

1 Introduction Gallium nitride (GaN) wide band-gap semiconductors have attracted much interest for optoelectronic and electronic applications. Although remarkable breakthroughs have been made for AlGaN/GaN high electron mobility transistors (HEMTs) [1-3], blue laser diodes, high-brightness light emitting diodes, and ultraviolet photodetector in the past decade, there still remains a bottleneck in the crystalline quality of GaN. According to the state-of-the-art of GaN epitaxial growth technology, most of the GaN epilayers are grown on c-plane sapphire substrate by metal organic chemical vapor deposition (MOCVD). Due to large lattice mismatch (16%) and thermal expansion coefficient difference between GaN and sapphire substrate, it is difficult to grow high quality GaN films with the threading dislocations (TDs) density less than ~10⁸ cm⁻².

To obtain high quality GaN epilayers on c-plane sapphire substrate by MOCVD, several kinds of interlayers are usually employed, such as AlGaN interlayer [4], low temperature AlN interlayer [5], and high temperature GaN interlayer [6]. AlN interlayer has been studied in detail for GaN growth on silicon substrate. Using multiple LT-AlN interlayers is an effective method to reduce the stress and to improve the crystal quality of GaN epilayers [7]. However, high temperature AlN interlayers have been little used for GaN growth on sapphire substrate. In this work, the emphasis is put on the effect of the growth temperature of AlN interlayer on the properties of the GaN epilayers. The related GaN epilayers were grown by MOCVD, and the AlN interlayers of different temperature (700 °C and 1050 °C) were designed which are labeled as low temperature AlN (LT-AlN) and high temperature interlayer (HT-AlN), respectively. The GaN epilayers were evaluated by high resolution X-ray diffraction (HRXRD) and Raman spectroscopy.

2 Experimental details The schematic diagrams of GaN epilayers with AlN interlayer are shown in Fig. 1, where the GaN epilayers were grown on c-plane sapphire substrate using a home-made MOCVD system. Trimethylgallium (TMG), trimethylaluminum (TMA), and ammonia (NH₃) were used as Ga, Al, and N sources, respectively. Hydrogen was used as carrier gas. The samples were grown starting with a low temperature AlN seed layer of
about 10 nm thickness at 550 °C followed by a 500 nm GaN buffer layer grown at 1000 °C. After that a 20 nm thick AlN interlayer was deposited prior to the growth of a 1000 nm thick top GaN epilayers at 1000 °C. The temperatures of AlN interlayer were 700 °C for the LT-AlN and 1050 °C for the HT-AlN samples, respectively.

3 Results and discussion

The crystalline quality of GaN epilayers was determined by high resolution X-ray diffraction (HRXRD) rocking curve measurements (Bruker D8 Discover HRXRD diffractometer). Additionally, Raman spectroscopy was used to investigate the residual stress in GaN epilayers using a 488 nm line from the argon ion laser with Z(XX)-Z scattering geometry at room temperature. The surface morphologies of GaN epilayers were characterized by atomic force microscopy (AFM).

The comparison of the FWHM values of GaN (0002) and (10-12) diffraction peaks determined in HRXRD measurements is shown in Fig. 2. The FWHM values of (0002) and (10-12) diffractions decrease from 478 arcsec and 1263 arcsec to 383 arcsec and 707 arcsec, respectively, when the temperature of AlN interlayer increases from 700 °C to 1050 °C.

It is well known that the FWHM values of rocking curves reflect the crystalline quality of GaN epilayers, and values of (0002) and (10-12) correspond to screw type and edge type dislocations, respectively. We can estimate the threading dislocation density by

$$D = \frac{\beta^2}{4b^2},$$

where $D$ is the density of threading dislocations, $\beta$ is the values of FWHM of rocking curve, and $b$ is the Burger vector. The values of $b$ for screw and edge dislocations have been given as 0.3189 nm and 0.5185 nm, respectively [8]. Thus the densities of screw and edge dislocations of GaN films with a HT-AlN interlayer are calculated to be $1.4 \times 10^8$ cm$^{-2}$ and $2.4 \times 10^8$ cm$^{-2}$, respectively. However, the values for GaN with a LT-AlN interlayer are $2.2 \times 10^8$ cm$^{-2}$ and $3.0 \times 10^8$ cm$^{-2}$, respectively. It indicates that the crystalline quality is improved significantly by using the HT-AlN interlayer compared with the LT-AlN interlayer. Obviously the threading dislocation propagation can be intercepted by the HT-AlN interlayer especially for edge type dislocations.

To further study the effect of temperature of the AlN interlayer on the residual stress in GaN films, Raman spectroscopy measurements were conducted. Figure 3 shows the Raman spectra of $E_2$ phonons of GaN epilayers with LT- and HT-AlN interlayers. The Raman shifts of $E_2$ phonons are 570.24 cm$^{-1}$ and 571.45 cm$^{-1}$ for GaN epilayers with LT- and HT-AlN interlayers, respectively. The Raman shift of $E_2$ phonons in free standing bulk GaN films is 567.5 cm$^{-1}$. The shift towards higher frequency suggests
that the epilayers are under compressive stress \cite{9}. Compressive stress exists in the GaN epilayers with HT- and LT-AlN interlayers, where the GaN epilayers with HT-AlN interlayer are under larger compressive stress.

In order to interpret the reason why the GaN crystal quality changes with AlN interlayer growth temperature, we need to analyze the growth mechanism of GaN films. Considering the fact that GaN films are formed by the coalescence of the crystalline grains during the GaN growth at high temperature, and the TDs are mainly located at the zones of coalescence \cite{10}, we believe that the temperature of AlN interlayer can account for the quality of films and the distribution of TDs. The AlN interlayer provides nucleation islands for the subsequent GaN growth. After growing in 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 quasi-2D mode.

The atomic force microscopy (AFM) images depicted in Fig. 4 show that the density of dark spots on the surface of the GaN sample with HT-AlN interlayer is much lower compared to that with LT-AlN interlayer. The dark spots are generated by dislocations and their lower number indicates that the density of threading dislocations is reduced by the HT-AlN interlayer, confirming the XRD results. The reason for the dislocation density reduction using HT-AlN interlayer is, however, not clear and needs to be further investigated.

\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{figure4.png}
\caption{AFM surface morphologies with a 5x5 \( \mu \text{m}^2 \) scan area of samples with (a) LT-AlN and (b) HT-AlN interlayer.}
\end{figure}

The strain in GaN epilayers is influenced by the initial island density and the annihilation of dislocations during layer growth \cite{11}. Most likely in GaN epilayers with a HT-AlN interlayer the compressive stress is higher than with a LT-AlN interlayer due to both a low density of GaN islands nucleating on the AlN layer and a more efficient annihilation of threading dislocations.

4 Conclusions The effect of the growth temperature of AlN interlayers on the properties of GaN epilayers grown on c-plane sapphire substrate by MOCVD has been studied. Based on the measurement and analysis, it is concluded that the crystalline quality of GaN epilayers is improved significantly by using HT-AlN interlayer, which can suppress the threading dislocation propagation to the GaN surface especially for edge type dislocations. The compressive stress was higher in GaN with a HT-AlN interlayer compared to films with LT-AlN interlayer.

Acknowledgements This work was supported by the Key Project of National Natural Science Fund of China (No. 60736033), the National Natural Science Fund of China (No. 60676048). The authors are grateful to Z. Li and S. F. Liu (Horiba JOBIN YVON Company in China) for Raman measurements.

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