Thin Solid Films 519 (2011) 5090–5094

Contents lists available at ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

Epitaxial growth of non-polar a-plane AlN films by low temperature sputtering using ZnO buffer layers

Hou-Guang Chen [a,*], Sheng-Rui Jian [a], Hui-Ling Kao [b], Meei-Ru Chen [b], Gou-Zhi Huang [a]

[a] Department of Materials Science and Engineering, I-Shou University, Kaohsiung 84001, Taiwan
[b] Department of Electronic Engineering, Chung Yuan Christian University, Chung-Li 32023, Taiwan

ARTICLE INFO

Available online 20 January 2011

Keywords:
ZnO
AlN
Epitaxial growth
Sputtering
MOCVD

ABSTRACT

This article presents an investigation on the epitaxial growth of non-polar a-plane AlN thin films by low temperature sputtering using ZnO buffer layers. Prior to the deposition of the AlN films, epitaxial growth of a-plane ZnO thin films on r-plane sapphire substrates was performed by a metalorganic chemical vapor deposition (MOCVD). The effect of MOCVD growth conditions on surface morphology and crystallinity of ZnO epi-layer was examined to optimize the growth process of buffer layer. The resulting ZnO epi-layers were used as buffer layers to grow non-polar AlN by low temperature sputtering. The measurements of XRD 2theta/omega- and phi-scans indicate that the epitaxial relationship among AlN, ZnO and sapphire substrate is $\left(11\bar{2}0\right)_{AlN}//\left(11\bar{2}0\right)_{ZnO}//\left(1\bar{1}02\right)_{Al2O3}$ and $\left[1\bar{1}00\right]_{AlN}//\left[1\bar{1}00\right]_{ZnO}//\left[11\bar{2}0\right]_{Al2O3}$. Cross-sectional transmission electron microscopy (XTEM) revealed that no reactive intermediate layers formed at the interfacial region between AlN film and ZnO buffer layer. The a-plane ZnO layers can be used as lattice matched templates for epitaxial growth of non-polar AlN by low temperature sputtering.

1. Introduction

Aluminum nitride (AlN) has been regarded as a candidate for applications in deep-ultraviolet (UV) light emitting diodes or laser diodes, and solar-blind photodetectors [1,2], due to its wide band gap of 6.2 eV and high thermal conductivity of 285 W/mK. In general, the performance of most AlN-based optoelectronic devices usually fabricated on polar (0001) plane suffers from the built-in polarization fields, resulting in the decrease of the carrier recombination rate in quantum well and degrading the quantum efficiency of devices [3,4]. The epitaxial growth on non-polar $\left(11\bar{2}0\right)$ a-plane, $\left(1\bar{1}00\right)$ m-plane or semi-polar $\left(11\bar{2}2\right)$ planes has been considered as an important solution to eliminating the impact of polarization fields [5,6]. Therefore, the investigation on the epitaxial growth of a- or m-plane AlN on various substrates, such as sapphire, SiC, and ZnO, has been devoted by numerous groups [7–9].

Sapphire is the most used as a substrate for epitaxial growth of III-group nitride films, owing to its low cost and availability in large size wafer. However, the epitaxial growth of non-polar AlN films on sapphire substrates with high crystalline quality is still a challenge, resulting from a large lattice mismatch between AlN and sapphire [7]. On the other hand, the use of ZnO epitaxial layers as underlying buffer layers for epitaxial growth of non-polar AlN layers on sapphire substrate is quite promising, because ZnO has the same crystallographic structure and close lattice matching with AlN. Furthermore, ZnO buffer layer can be served as a sacrificial layer for fabricating free-standing single crystalline templates of GaN or AlN through chemical lift-off process [10]. However, the problem emerges from epitaxial growth of III-group nitride films on ZnO surfaces, since the formation of reactive layers at interface between III-group nitride and ZnO at temperatures above 500 °C negates the advantages of lattice-matched template [11]. Therefore, low temperature physical vapor deposition (LT-PVD) technique, such as room temperature pulsed laser deposition, have been developed for epitaxial growth of AlN on ZnO substrates to overcome the problems of the formation of interfacial layers [9]. Recently, the low temperature (300 °C) epitaxial growth of high quality AlN layers on GaN templates using the helicon sputtering system have been reported by Kao et al [2,12]. In helicon plasma system, the sputtered species were ionized efficiently by inductively coupled radio frequency plasma. Highly efficient ionization of sputtered species permitted epitaxial growth of AlN films on lattice-matched substrates at a relatively low temperature. In this study, the microstructure of non-polar a-plane AlN thin films deposited by low temperature helicon sputtering system on MOCVD grown ZnO buffer layers has been investigated by X-ray diffraction (XRD) and cross-sectional transmission electron microscopy (XTEM) techniques. In addition, the effect of MOCVD growth conditions on surface morphology and crystalline quality of a-plane ZnO buffer layers was also demonstrated in this work.

* Corresponding author. Tel.: +886 7 6577711x3113; fax: +886 7 6578444.
E-mail address: houguang@isu.edu.tw (H.-G. Chen).

0040-6090/$ – see front matter
doi:10.1016/j.tsf.2011.01.149

2. Experimental procedure

Prior to AlN deposition, the epitaxial growth of a-plane ZnO buffer layers on $1\times1\ cm^2$ $(1\bar{1}02)$ r-plane sapphire substrates was implemented by a reduced pressure hot-wall type MOCVD apparatus. Zinc acetylacetonate ($Zn(acac)_2$, 99.995 purity) and O_2 (99.999 purity) were used as the zinc and oxygen source, respectively. The precursor of $Zn(acac)_2$ was heated at 100 °C before its introduction into the chamber by the carrier gas of nitrogen. The ZnO thin films were grown at temperatures of 470–550 °C for 120 min, and the chamber pressure was kept at 120 Torr. The AlN films were deposited on ZnO buffer layers by using a helicon sputtering system. An aluminum target (99.999% purity) having two inches in diameter was placed at a cathode, and the argon and nitrogen mixture gas (Ar:N = 3:1) was introduced with a constant pressure of 1 mTorr. During the deposition of AlN, the RF power was operated at 150 W and the coil power was 50 W. The effect of substrate temperature on the growth of AlN films on ZnO buffer layers was examined. Microstructure of the epitaxial film was characterized by using X-ray diffraction (XRD, Philips X'Pert diffractometer with a Cu K_α radiation source). The scanning electron microscopy (FESEM, Hitachi S-4700) was used to examine the morphologies of the films. Cross-sectional TEM observation was performed on a Philips Tecnai 20 microscope operated at 200 kV.

3. Results and discussions

The epitaxial growth of a-plane ZnO buffer layers on r-plane sapphire substrates have been carried out by MOCVD at temperatures of 470, 500, and 550 °C, respectively. The surface morphologies of ZnO buffer layers grown at various temperatures were observed by SEM. The growth temperature has been found significantly to affect the surface morphology of ZnO layers. In Fig. 1(a), the characteristic feature on the surface of ZnO film grown at 470 °C is the triangular shaped pits with a density of $3.75\times10^{10}\ cm^{-2}$. The similar pitted defects which decorate threading dislocation terminations at surface has been commonly reported for a-plane GaN epitaxially grown on r-plane sapphire substrates. The in-plane asymmetric growth rate on the a-plane of the wurtzite structure might be responsible for the formation of the triangular-shaped pits on the ZnO or GaN surfaces [13]. Remarkably, the heavily pitted surface morphologies disappear with further increasing growth temperature above 500 °C, as shown in Fig. 1(b) and(c). The epitaxial relationship between ZnO and sapphire has been established to be $(11\bar{2}0)_{ZnO}//(1\bar{1}02)_{Al2O3}$ and $[1\bar{1}00]_{ZnO}//[11\bar{2}0]_{Al2O3}$, as shown elsewhere [14]. The crystallinity was evaluated from the full width at half maximum (FWHM) values of X-ray rocking curve (XRC) for $(11\bar{2}0)$ reflection. For non-polar a-plane ZnO epitaxy on r-plane sapphire, the calculated in-plane lattice mismatches are 1.5% for the $[0001]_{ZnO}//[\bar{1}101]_{Al2O3}$ and 18% for the $[1\bar{1}00]_{ZnO}//[11\bar{2}0]_{Al2O3}$, respectively; hence, the presence of in-plane anisotropic mosaicity in the a-plane ZnO epitaxy on r-plane sapphire is expectable, due to in-plane anisotropic strain relaxation [15,16]. Therefore, in order to characterize the in-plane anisotropic mosaicity, the XRC measurements were performed at two different azimuth angles (φ), where the projection of X-ray incident beam is parallel to the ZnO [0001] direction (φ = 0°), or to the ZnO $[1\bar{1}00]$ direction (φ = 90°). The variation in the FWHM of $(11\bar{2}0)$ XRC as a function of the growth temperature is demonstrated in Fig. 1(d). The reduction in XRC FWHM as increasing growth temperature possibly results from that the enhancement of adatoms diffusion on growing surface under higher temperature conditions is effective in improving

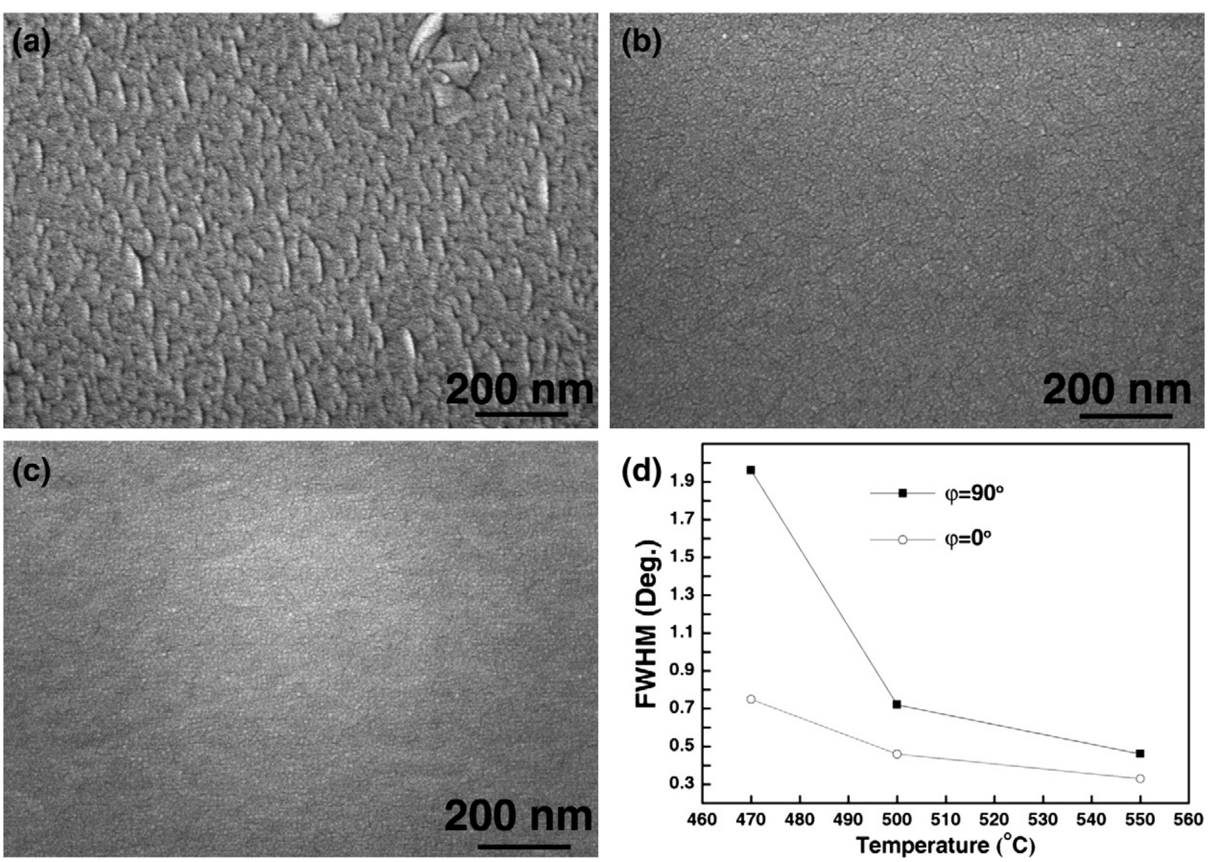

Fig. 1. The SEM images of the a-plane ZnO epitaxial films grown by MOCVD at temperatures of (a) 470 °C, (b) 500 °C, and (c) 550 °C, respectively. (d) The variation in the FWHM of $(11\bar{2}0)$ XRC recorded at different azimuth angles (φ) as a function of the growth temperature.