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Influence of ZnO buffer layer thickness on crystalline quality of ZnO thin film

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The effect of homo-buffer layer thickness on the crystalline quality and the surface roughness of ZnO thin film was investigated. ZnO thin films were prepared on (0001) sapphire substrates with different homo-buffer layer thickness of 20~60 nm by a radio-frequency magnetron sputtering method. The buffer layers were deposited at a substrate temperature of 400 °C and then main ZnO films were grown on the buffer layers at 600 °C. For comparison, single-layered ZnO films without buffer layer were prepared at substrate temperatures of 400 °C and 600 °C, respectively. Compared to the films without buffer layer, the films with buffer layer exhibited higher crystalline quality and surface smoothness. For the ZnO films with buffer layer, the transmittance spectra exhibited a steep fall-off at 380 nm, which is a characteristic of high quality ZnO film. The highest ZnO film quality was obtained for the film prepared with a buffer layer thickness of 20 nm, from which it is suggested that the crystalline quality and surface smoothness of ZnO film are improved as the buffer layer thickness decreases.

Key words: ZnO thin film, Homo-buffer layer, Buffer layer thickness, Crystalline quality.

Introduction

Wide band gap semiconductors such as GaN and ZnO have been intensively studied during the past several years because of their applications in optoelectronic and microelectronic devices such as light emitting diodes and laser diodes. Particularly, ZnO has a direct bandgap of 3.37 eV and a large exciton binding energy of 60 meV. Its binding energy is much larger than that of GaN (24 meV) and the thermal energy at room temperature (26 meV), which results in an efficient excitonic lasing even at room temperature under low threshold voltage [1, 2]. Thus ZnO has been paid much attention for applications in photonic devices in the UV region. In addition, ZnO shows the inherent n-type characteristic, which is caused by high concentration of free electron originated from intrinsic internal defects such as oxygen vacancies and zinc interstitials. This makes it one of the most promising materials for transparent conductive films suitable for the front electrode in thin film solar cell and flat panel display [3, 4]. It is well known that the optical and electronic properties of ZnO films are significantly dependent on the crystalline quality of the films. Therefore to assure the high performance of ZnO-based devices, it is very important to fabricate ZnO films with high crystalline quality and smooth surface. Therefore much effort has been made to obtain ZnO films with high crystalline quality.

Many techniques have been used to grow ZnO films with high crystalline quality. Among the growth techniques, the technique to use buffer layer is very effective for the improvement of the crystalline quality of ZnO films. So far, a variety of buffer layers such as CaO [5], SiC [6], MgO [7] have been employed as buffer layers to improve the crystalline quality and optoelectronic performances of ZnO films. Most recently, it was reported that low temperature grown ZnO homo-buffer layer plays an important role in the improvement of the crystalline quality of ZnO films [8, 9].

This paper reports the effect of ZnO buffer layer thickness on the crystalline quality and surface smoothness of ZnO film deposited on (0001) sapphire substrate by a RF magnetron sputtering.

Experimental

ZnO buffer layers and main ZnO films were sequentially deposited by radio-frequency (RF) magnetron sputtering method. Sapphire with (0001) orientation was used as a substrate. Prior to deposition, the substrate was ultrasonically cleaned with acetone and ethanol, and then rinsed with deionized water. After the substrate being loaded into the sputtering chamber, the sputtering chamber was evacuated to 5.0×10^{-6} torr by a turbo-molecular pump before deposition. A sintered ZnO specimen with a purity of 99.99% was used as a target. Ar was used as gas for sputtering. Deposition was carried out at a constant working pressure of 2.5×10^{-3} torr and an RF power of 150 W. Before the deposition, the target was pre-sputtered for 10 min to remove any contamination. The substrate holder was 10 cm away from the target and was rotated for the improvement on

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the uniformity of film thickness. The homo-buffer layer was deposited at a substrate temperature of 400 °C and the deposition of the main ZnO film was performed at 600 °C. To investigate the effect of buffer layer thickness on the film quality, the buffer layer thickness was changed in range of 20~60 nm. Then the ZnO films were grown on the buffer layers with different thicknesses. Total thickness of buffer layer and main film in all samples was estimated to be about 200 nm. For comparison, the samples with the same thickness of 200 nm were also prepared without buffer layer at constant temperatures of 400 °C and 600 °C, respectively. Five samples were prepared. Sample (a) and (b) were prepared with a thickness of 200 nm through single-step process at 400 °C and 600 °C, respectively. Sample (c), (d) and (e) were prepared with different buffer layer thicknesses of 20, 40 and 60 nm, respectively. The sample (c) was composed of 20 nm thick buffer layer and 180 nm thick main film. In case of the sample (d), the thicknesses of the buffer layer and main film were 40 and 160 nm, respectively. The sample (e) was prepared with a buffer thickness of 60 nm and a main film thickness of 140 nm.

The crystalline structures of the ZnO films were investigated by X-ray diffractometer using Cu-K_α radiation operated at 40 kV and 30 mA. The surface morphology of the films was observed by scanning electron microscopy (SEM) and atomic force microscopy (AFM). Optical transmittance was measured by a UV-Vis-NIR spectrophotometer in the wavelength range of 200-600 nm, from which the optical band gaps of the films were determined.

Results and discussion

Fig. 1 shows the XRD spectra of the samples. The diffraction patterns for the samples can be well indexed to hexagonal wurtzite structure of ZnO. The diffraction peaks from only ZnO (002) and (004) planes were observed in the spectra of the samples, indicating that

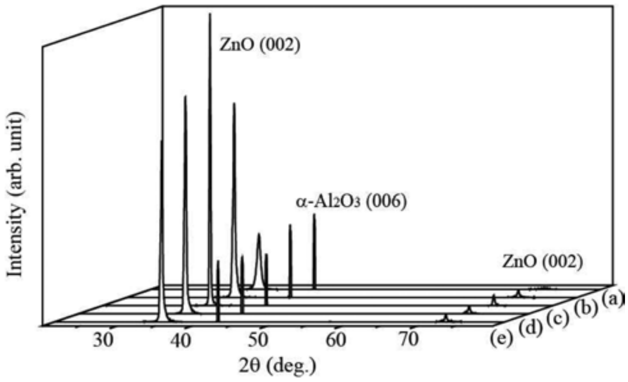

Fig. 1. XRD patterns of the ZnO films with and without buffer layer.

the ZnO films have hexagonal wurtzite structure with highly c-axis orientation perpendicular to the substrate. The intensity of (002) peaks in the spectra of the ZnO films with buffer layers is stronger than that for the ZnO films without buffer layer, which means that the introduction of homo-buffer layer is very effective for the improvement in crystalline quality of ZnO thin film. The (002) peak intensity of the sample with the buffer layer thickness of 20 nm is the strongest among the samples with buffer layers, indicating that the buffer layer thickness has an effect on the crystallization of ZnO film. The full-width at half-maximum (FWHM) of the (002) diffraction peaks for the samples (a), (b), (c), (d) and (e) was estimated to be 0.58°, 0.28°, 0.20°, 0.25° and 0.28°, respectively. The smallest FWHM is founded for the sample with the buffer thickness of 20 nm, indicating that the ZnO film deposited on 20 nm thick buffer layer has the best crystalline quality. This result also suggests that the crystalline quality of ZnO film is significantly influenced by the buffer layer thickness. From the XRD results, it is concluded that high-quality ZnO film can be achieved by introducing the low-temperature ZnO homo-buffer layer, and the buffer layer thickness affects the crystalline quality of ZnO main film.

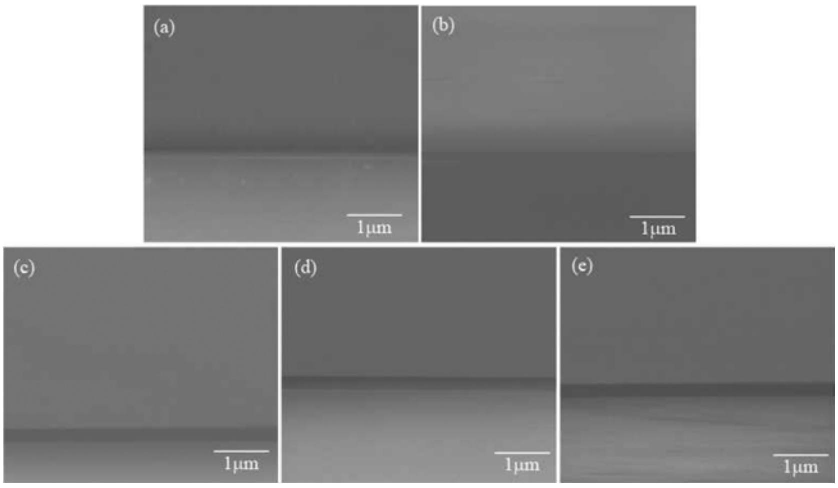

Fig. 2. SEM micrographs of the ZnO films with and without buffer layer.

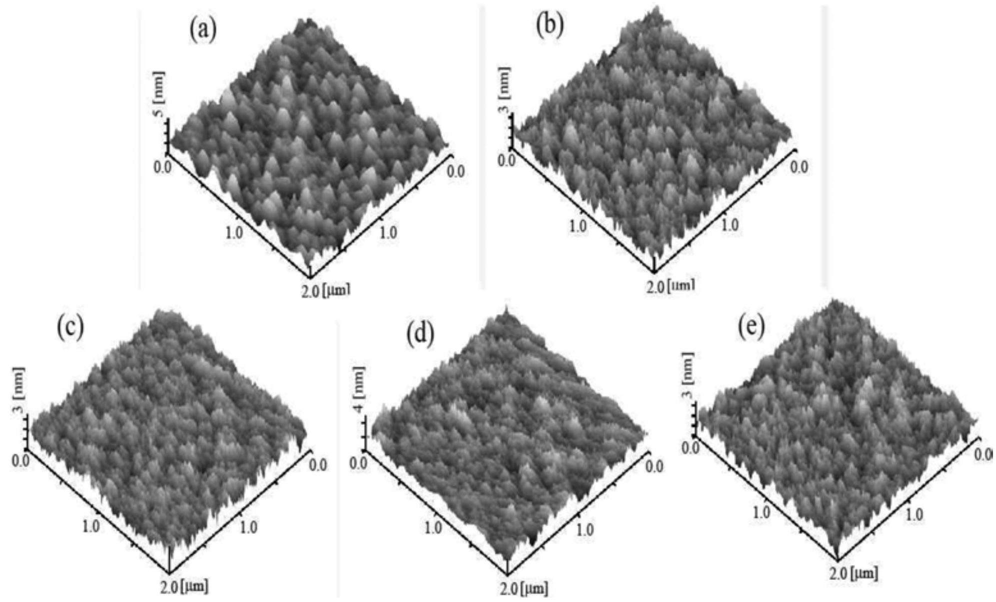

Fig. 3. AFM images measured from the ZnO films with and without buffer layer.

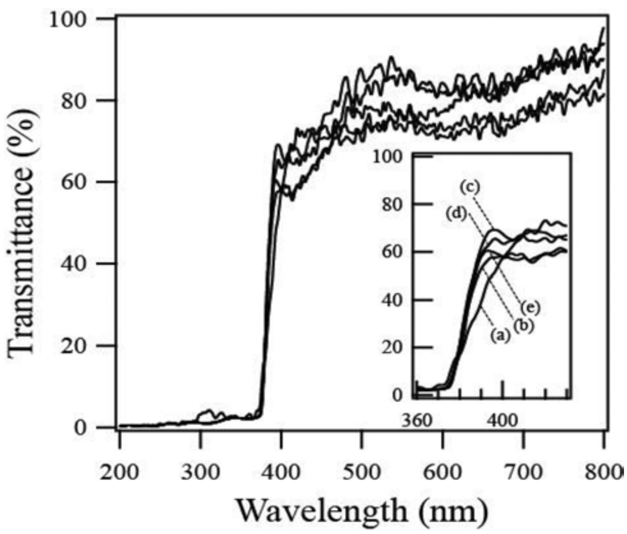

Fig. 4. Optical transmittance spectra of the ZnO films prepared with and without buffer layer.

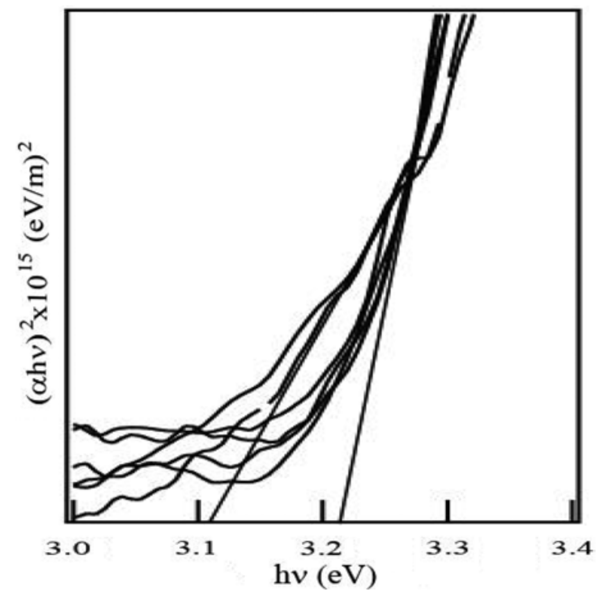

Fig. 5. Plots of $(ahv)^2$ vs hv of the ZnO films prepared with and without buffer layer.

Fig. 2 shows SEM micrographs taken for the samples (a), (b), (c), (d) and (e). The surface with flat and morror-like smoothness is observed for all samples regardless of whether or not buffer layer was used. To investigate the surface morphology of the ZnO films in more detail, AFM measurement was performed for the samples.

Fig. 3 represents AFM images measured from the samples (a), (b), (c), (d) and (e). The AFM images were taken on an area of 1×1 m^2. The surface roughness was estimated as root-mean-square (RMS) of a surface profile. The RMS values measured from AFM images of the samples (a), (b), (c), (d) and (e) are 1.047, 0.421, 0.395, 0.412 and 0.475 nm, respectively. The sample (c) with 20 nm thick buffer layer shows the smoothest surface. As the buffer layer thickness increases further, surface morphology of ZnO films becomes rough, which is supposed to be ascribed to the increase of nucleus size. The nuclei get larger as buffer layer thickness increases. When atoms adsorb on the top of the large nuclei, the atoms cannot fall easily down to lower level from the top of the nuclei. This results in the formation of new nuclei on the pre-existing nuclei, leading to the enhancement in three-dimensional growth. Consequently, the surface roughness of the film increases with increasing the buffer layer thickness.

Transmittance and optical band gap were also estimated for evaluating the crystalline quality of ZnO films and shown in Fig. 4. All the ZnO films show high transmittance over 70% in the wavelength range of visible light. However, as shown in the inset of Fig. 4, the fall-off near absorption edge in transmittance spectra of the samples with buffer layer was sharper than those of the samples without buffer layer, which indicates that the crystalline quality of ZnO film is improved by using ZnO buffer layer. The optical band gaps were also estimated to be 3.12, 3.12, 3.22, 3.22 and 3.22 eV for the sample (a), (b), (c), (d) and (e) as shown in Fig. 5. The band gaps were calculated by plotting $(\alpha hv)^2$ vs hv from the transmittance spectra using an $\alpha hv = A(hv\text{-}Eg)^{1/2}$ relationship, in which α is the absorption coefficient and hv is the photon energy, and extrapolating the straight line portion of this plot to the energy axis. For stoichiometric ZnO film, the optical band gap is known to be 3.24 eV [10]. Therefore, the result reveals that the ZnO films prepared on buffer layers have high crystalline quality.

Conclusions

ZnO films without and with homo-buffer layer were deposited on sapphire (0001) substrates by RF magnetron sputtering method. All films had wurtzite structure and were grown with c-axis orientation perpendicular to the substrate. Compared with ZnO films without buffer layer, ZnO films with buffer layer had higher crystalline quality and smoother surface. In addition, as the thickness of buffer layer decreased, the crystalline quality and surface smoothness of ZnO film were significantly improved. ZnO film deposited on buffer layer with a thickness of 20 nm exhibited the highest crystalline quality and the smoothest film surface. From these results, it can be concluded that buffer layer thickness is one of the key factors to fabricate ZnO thin film with high crystalline quality and smooth surface.

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