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Effect of Growth Temperature on Structural and Electronic Properties of ZnO Thin Films

Dahlang Tahir[1,a)] and Kang Hee Jae[2]

[1]*Department of Physics, Hasanuddin University, Makassar, 90245 Indonesia*
[2]*Department of Physics, Chungbuk National University, Cheongju, 361-763 KOREA.*

[a)]Corresponding author: dtahir@fmipa.unhas.ac.id

Abstract. The electronic and structural properties for RF magnetron sputtering deposited ZnO thin films grown on Si substrate was obtained by using X-ray diffraction (XRD) and reflection electron energy loss spectroscopy (REELS). XRD spectra show the intensity of the diffraction peak increases with increasing the growth temperature with (002) is strongest diffraction peak. The particle sizes for (002) are increase from 10.2 nm to 60.2 nm with increasing growth temperature from room temperature to 500°C, respectively. The band gap of ZnO thin films REELS spectra are 3.1±0.1 eV. The dominant peak from REELS at about 18 eV is ascribed to a bulk Plasmon excitation, which represents the collective oscillation of the valence electrons excited by the incident fast electrons.

21 April 2024 10:33:52

INTRODUCTIONS

Zinc Oxide (ZnO) has attracted increasing interest because of its superior electronic and optical properties leading to a wide range application such as organic and amorphous semiconductor for plastic electronic, light-emitting diodes, photodetector, transparent conducting films for solar cells, and transparent thin film transistors [1]. Band structure and optical properties of ZnO are very similar to those of GaN, which is known to be good materials for optical devices such as light-emitting diodes or laser diodes [1,2]. ZnO normally has the hexagonal structure with a= 3.25Å and *c*= 5.12 Å; each Zn atom is tetrahedrally coordinated to four O atoms, where the Zn d-electrons hybridize with the O p-electrons; layers occupied by zinc atoms alternate with layers occupied by oxygen atoms. Several techniques can be used to grow ZnO thin films on variety of substrate (Si, glass, flexible substrate, and sapphire), such as spray pyrolysis, Magnetron sputtering, reactive plasma deposition, metal organic chemical vapor deposition, and pulse laser deposition [1-5].

There are many studied have been investigate on structural, electronic and optical properties with varying dopant concentration and also process and treatment after deposition to obtain high quality and performance devices based ZnO thin films. The structural, electrical and optical properties of ZnO significantly improve with high dopant concentration and after treated by argon, oxygen, Nitrogen, and chlorine plasma. In additions, hydrocarbon impurities in ZnO inevitably incorporated during deposition have negative effect on device performance [1,4,6].

The structural, electronic and dielectric properties of ZnO thin films are essential in understanding the transport properties transparent conducting solar cells devices based ZnO. So far, effect of growth temperature on structural and electronic properties of ZnO thin films deposited on Si substrate have been rarely experimentally investigated. Therefore, in this work we studied the effect of growth temperature on structural from XRD data and the electronic properties at room temperature from REELS measurement as a key for electrical properties.

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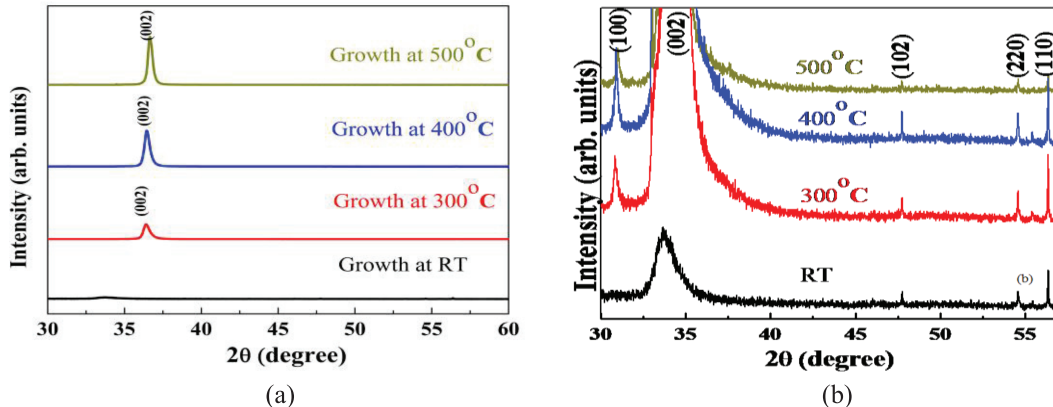
EXPERIMENT

ZnO thin films were deposited by rf magnetron sputtering on Si substrates for the rf power of 200 W with variety growth temperature; room temperature, 300°C, 400°C and 500°C in the argon gas containing 65% of oxygen. Structural properties were determined by using XRD data. XRD data was obtained from the performance a Philips PW3710 for scan 2θ from 20 to 60. REELS spectra were obtained by using the VG ESCALAB 210. Spectra were measured using Al source with the pass energy of 20 eV. The incident and take off angles of electrons were 55° and 0° from the surface normal, respectively. REELS were measured with the primary electron energies of 1.0 and 1.5 keV for excitation and with the constant analyzer pass energy of 20 eV. The full width at half maximum (FWHM) of the elastic peak was 0.8 eV.

RESULTS AND DISCUSSION

X-ray diffraction spectra of the doped ZnO thin films are shown in Fig. 1. X-ray diffraction spectra of the films were taken with different growth temperature. It has been observed that the XRD peak broadening decreases with increase of growth temperature. The peaks with the Miller indices given belong to the ZnO presence of five peaks with orientations; (100), (002), (102), (220) and (110) confirmed these X-ray diffraction patterns are polycrystalline for ZnO thin films deposited on silicon [7]. The intensity of these diffraction increases with increasing the growth temperature. The preferred orientation is (002) as indicated by the strongest diffraction peak for all growth condition as we can see clearly in Fig. 1.(a). Other peaks as indicated in Fig. 1.(b) with much less intensity was observed. The highest peak value of the XRD measurement was obtained from the ZnO thin films deposited at 500°C, indicated shows the best crystal structure [2]. It was observed that the crystallite size for the highest intensity increased with increasing growth temperature as shown in Table 1.



(a) (b)

FIGURE 1. XRD pattern for ZnO thin films with various growth temperatures (a) the strongest diffraction pattern (b) all diffraction pattern (zoom in figure 1(a)).

The grain size of thin films ZnO were determined from the XRD spectra by using Scherrer's formula $d = \frac{0.9\lambda}{B\cos\theta}$, where λ, θ and *B* are the X-ray wavelength (1.54 Å), Bragg diffraction angle and line of full width at half maximum, respectively [7]. It was determined that there is a continuous increase in particle size with increasing growth temperature. To examine the quality of the thin films ZnO for different growth condition, the full width at half maximum (FWHM) of (002) peak and the crystallite dimension are estimated. The FWHM values for ZnO on Si are 1.41, 0.33, 0.26 and 0.24 for growth temperature at room temperature, 300°C, 400°C and 500°C, respectively. It is indicated the growth temperature above room temperature exhibiting good crystallinity. The films has the highest peak value of XRD and the narrowest value of FWHM, it has the best crystal quality [6,7]. The particle size of thin films ZnO for (002) was determined according to Scherrer in a around 10.2 nm at room temperature and increased to 60.2 nm for the growth temperature 500°C. The increasing particle size is due to the emerging of the smaller particles into larger ones resulting from the potential energy difference between small and large particles, which was occur through solid state diffusion [8].

21 April 2024 10:33:52

TABLE 1. Full width at half maximum (FWHM) and particle size of ZnO thin films for (002) diffraction pattern.

No.	Growth Temperature	FWHM	Particle Size (nm)
1	RT	1.41	10.2
2	300°C	0.33	43.7
3	400°C	0.26	55.9
4	500°C	0.24	60.2

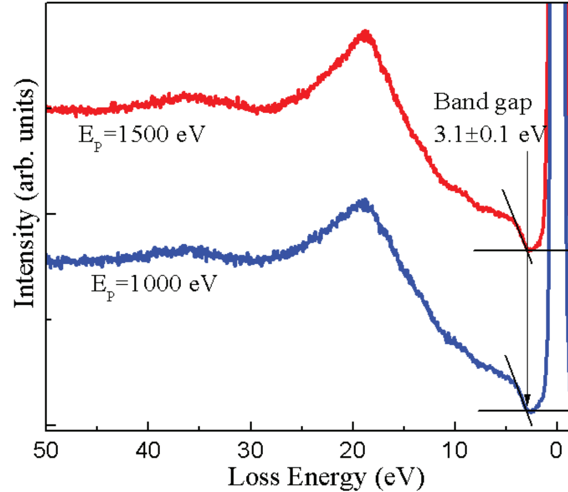


FIGURE 2. REELS spectra of ZnO thin films for primary energies of 1.0 and 1.5 keV.

21 April 2024 10:33:52

Figure 2 shows the REELS spectra for the ZnO on Si substrate thin films at the primary energy of 1.0 and 1.5 keV. The onset of a loss is due to electron-hole excitation and corresponds to the band gap value of ZnO thin films. The band gap energy can thus be found by drawing a linier fit line with a maximum negative slope from a point near the onset of the loss spectrum to the background level. The crossing point gives the band gap value. The band gap of ZnO thin films are 3.1±0.1 eV. This band gap is lower than that of crystalline ZnO reported by other references (3.37 eV) [1]. The method was described in our previous paper [8-11]. Several common features are observed in REELS spectrum; a small peak at 9.5 eV and 13.5 eV, dominant peak at 18.8 eV, 21.8 eV and another shoulder peak at 36.5 eV away from elastic peak.

In the loss function of ZnO thin films, the dominant peak at about 18 eV is ascribed to a bulk Plasmon excitation, which represents the collective oscillation of the valence electrons excited by the incident fast electrons. Several other common peaks at 9.5, 13.5, and 21.8 eV are identified respectively as interband transition from the O *2p*, the Zn *3d*, and the O *2s* states in the valence band to occupied states in the conduction band. These peaks position agree well with those of nano-wire ZnO, indicating that the general electronic structure of the ZnO thin films are in nano dimension [12,13].

CONCLUSION

In summary, we investigated the structural and electronic properties of ZnO thin films deposited on Si substrates via XRD and REELS analysis. XRD spectra indicated multi-crystalline present for ZnO deposited on Si. The ZnO thin films for growth temperature 500ºC the best crystal quality, indicated by the highest peak value of XRD and the narrowest value of FWHM. The crystallite size of thin films ZnO for (002) was around 10.2 nm at room temperature and increased to 60.2 nm for the growth temperature 500ºC. The optical band gap of the ZnO thin films deposited at room temperature are about 3.1 eV. In the loss energy spectrum, the dominant peak at about 18 eV is ascribed to a bulk Plasmon excitation. Peaks at 9.5, 13.5, and 21.8 eV are interband transition from the O *2p*, the Zn *3d*, and the O *2s* states in the valence band to occupied states in the conduction band.

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21 April 2024 10:33:52