Fabrication and characterization of ZnO/Al/ZnO multilayers by simultaneous DC and RF magnetron sputtering

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Abstract. The present investigation reports the fabrication and characterization of multilayered transparent electrodes by simultaneous DC and RF magnetron sputtering on glass substrates. The multilayer structure consists of three layers (ZnO/Al/ZnO). The influence of Al layer thickness on the electrical and optical properties was investigated. Optimum thickness of Al was determined for high transmittance and good electrical conductivity. High quality films having resistance as low as 25 Ω/sq with optical transmittance upto 65% were obtained at room temperature.

1. Introduction

Transparent conducting oxides (TCOs) are widely used in many technological applications such as solar cells, optoelectronic devices, consumer electronics etc. Tin doped indium oxide (ITO) one of the most used TCOs at present. But, its toxicity and high cost made to look for other alternatives. Zinc oxide (ZnO) is one of the most promising materials for the fabrication of the next generation optoelectronic devices in the UV region and optical display devices. It has many advantages such as less toxicity, less cost and availability in the large scale. It is a direct band gap semiconductor having an energy gap of 3.37 eV at room temperature with 60 meV exciton binding energy [1-3]. It has been subjected to extensive research by the research community due to its attractive properties. The doping effect on its properties is still under investigation. Doped ZnO films have many interesting potential applications such as transparent electrodes, piezoelectric devices, gas sensors, photo diodes and also it can be used as window layer in hetero junction solar cells [4-6]. The main objective of developing new materials for TCO is to achieve less resistivity and good transparency in the visible range of the electromagnetic spectra.

Different methods were employed to develop good TCOs by many researchers. Multilayer stacks of TCOs are one such approach employed to improve the properties of TCOs [7-10]. Kawashima et al. reported improvement of thermal stability in F:SnO_{2}/ITO stack compared to single layer ITO [11]. Similar stacks of Sn: CdO/CdIn_{2}O_{4}/Cd_{2}SnO_{4} showed better conductivity than ITO with comparable transparency in the visible region [12]. The deposition of three layer structures with a metallic film embedded between two dielectric layers (D/M/D) exhibits better TCO properties with thickness less than a single layer TCO. These D/M/D structures are widely used in energy saving widows as optical filters [13]. High refractive index zinc oxide makes it a suitable candidate as a dielectric layer. Few early reports on D/M/D include the development of ZnO/Ag/ZnO & ITO/Ag/ITO [14], ITO/Cu/ITO [15], TiO_{2}/Ag/TiO_{2} [16] and ZnO/Cu/ZnO [17]. Ting et al. reported insertion of thin Al interlayer for the improvement of electrical conductivity of ZnO [18].
In the current work, we report the deposition of ZnO/Al/ZnO multilayered structure by simultaneous RF sputtering of ZnO and DC sputtering of Al targets at room temperature. We investigate the effect of Al layer on the properties of multilayers.

2. Experimental
Multilayers of ZnO/Al/ZnO were deposited on pre-cleaned glass substrates. Target materials are compacted zinc oxide (99.99% purity, 2 inch dia., 3 mm thick) and metallic Al (99.999% purity, 2 inch dia., 3 mm thick). The magnetron sputtering system consists of 12 inch vacuum chamber which is equipped with 2 inch magnetron cathodes and RF and DC power supplies. Prior to deposition chamber was pumped down to a pressure less than $5 \times 10^{-6}$ m bars. High pure argon was used as sputter gas and working pressure was adjusted at 0.001 m bars. Sputtering power was kept 150 Watts for ZnO and 15 Watts for Al. The optical characterizations of alms have been carried out using UV–Vis spectrophotometer (USB2000). Electrical resistance of the deposited multilayers was measured using point probe technique using Keithley 2400 sourcemeter and Keithely 2000 multimeters.

3. Results and discussion
X-ray diffraction patterns of the deposited multilayers are shown in Figure 1. The thickness of the ZnO layer was found around 100 nm and Al layer thickness varies from 5 nm to 30 nm. Thickness of Al layer was calculated by considering the deposition rate of Al. A single peak at $2\theta \approx 34.75^\circ$ confirms the existence of ZnO phase. It is also revealed that the deposited films show preferred orientation along (0 0 2) planes. The intensity of (0 0 2) peak found to decrease with the increase in Al layer thickness which indicates the decrease in the crystallinity. No prominent peaks corresponding to Al were observed. This may be because of the fact that the thickness of the ZnO layer is large compared to that of Al. The observed 20 positions for multilayer film was bit high compared to that of ZnO film studied here. This is because of residual stress shows optical transmittance spectra of ZnO/Al/ZnO multilayer with variation in Al deposition time.

![Figure 1. XRD patterns of ZnO and multilayers with different Al interlayer thickness](image)

Optical transmittance spectra of the deposited films are shown in Figure 2. For undoped films a good transmittance over 80% was observed in the visible region of the electromagnetic spectra. The average transmittance of the films in the visible region decreases as the Al layer thickness increases. Absorption edges of the multilayers shift towards higher wavelengths with increase in the thickness of Al interlayer thickness. As the thickness of Al interlayer increases the electrical resistance decreases (Figure 3). Multilayered thin films show low resistance because of Al interlayer. The conduction electrons were supplied from donor sites associated with excess zinc and aluminum ions on
substitutional sites of zinc ions in the in the as deposited samples. The lowest resistance obtained for the multilayer thin film with Al interlayer thickness 33 nm.

**Figure 2.** Transmittance spectra of ZnO and multilayer thin films.

**Figure 3.** The variation of sheet resistance and average transmittance (400-875 nm)

4. Conclusions

We deposited multilayers of ZnO/Al/ZnO were deposited by simultaneous RF and DC sputtering. Obtained multilayers were characterized using optical transmittance spectra and XRD. Sheet resistance of the multilayers found to decrease with the increase in the Al interlayer thickness. XRD patterns of the multilayers show reflections corresponding to Wurtzite ZnO, no peaks corresponding to Al observed due to thicker ZnO layers. Transmittance spectra show a considerable decrease in the average transmittance (in the visible region) of the multilayers.

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