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BIAS VOLTAGE DEPENDENCE PROPERTIES OF DC REACTIVE MAGNETRON SPUTTERED INDIUM OXIDE FILMS

P. Mohan Babu*, B. Radhakrishna[a], G. Venkata Rao, P. Sreedhara Reddy, S. Uthanna

Department of Physics, Sri Venkateswara University, Tirupati – 517 502, India
[a]Department of Physics, PBR Visvodaya Institute of Technology and Science, Kavali – 524 201, India

Thin films of indium oxide were deposited by dc reactive magnetron sputtering onto glass substrates held at various bias voltages and temperatures. The films formed without substrate biasing and substrate heating were amorphous. The films deposited with substrate heating and / or substrate biasing were polycrystalline with predominant (222) orientation. The influence of substrate bias on the structural, electrical and optical properties was systematically studied. The films formed at a substrate temperature of 473 K and substrate bias voltage of –100 V resulted in transparent conducting films with electrical resistivity of 1.8×10^{-3} Ω cm, optical transmittance of 85% and figure of merit of 1.1×10^{-3} Ω^{-1}.

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Keywords: Indium oxide films, Magnetron sputtering, Structure, Electrical and optical properties

1. Introduction

Indium oxide (In_2O_3) is n-type highly degenerate, wide band gap semiconductor which exhibits high electrical conductivity and high optical transparency in the visible light region [1]. These unique properties are extensively useful as transparent conductors in optoelectronic devices such as transparent electrodes for flat panel displays [2], selective transparent coatings for solar energy heat mirrors [3] and window layers in heterojunction solar cells [4, 5]. A wide range of thin film deposition techniques such as vacuum evaporation [6 – 8], thermal oxidation of indium films [9], pulsed laser deposition [10 – 12], atomic layer epitaxial growth [13], spin coating [14], sol-gel process [15], ion assisted deposition [16], and dc and rf sputtering [18 – 21] were employed for preparation of indium oxide films. In recent years much attention has been focussed on dc reactive magnetron sputtered In_2O_3 films because of their advantages such as sputtering from metallic indium target in the presence of reactive gas of oxygen, high deposition rates, film uniformity on large area and precise control over the composition of the deposited film. The physical properties of In_2O_3 films prepared by this method mainly depend on the sputtering parameters like oxygen partial pressure, substrate temperature, substrate biasing, sputtering pressure apart from the sputtering power and target to the substrate distance. The effect of substrate temperature [20] and oxygen partial pressure [22] on the physical properties of In_2O_3 films were reported earlier. In order to achieve high electrical conductivity and optical transparency the substrate should be maintained at elevated temperatures during deposition of the films. This prevents to use temperature sensitive substrates like plastics which is extensively used as substrates for optoelectronic devices [23]. The effect of substrate heating can also be achieved by ion bombardment on the growing film during deposition. The simplest way of achieving this is by biasing the substrate during the deposition of the films. In this investigation, we studied the influence of substrate bias on the crystallographic structure, electrical and optical properties of In_2O_3 films prepared by dc reactive magnetron sputtering.

* Corresponding author: gutlapalliv@yahoo.com

2. Experimental

In_2O_3 films were deposited on glass substrates by dc reactive magnetron sputtering method. The sputtering chamber was pumped to 1×10^{-5} mbar using diffusion pump and rotary pump combination. Pressure in the sputtering chamber was measured using digital Pirani-Penning gauge combination. A circular planar magnetron of 100 mm diameter was used as the magnetron cathode. The magnetron target assembly was mounted on the top plate of the sputtering chamber so that the sputtering can be achieved in the sputter down mode. A continuously variable dc power supply of 750 V and 3 A was used a power source for sputtering. Indium (99.995% pure) target was used as sputtering target. Pure argon and oxygen gases were used as sputtering and reactive gases respectively. The flow rates of both the gases were controlled individually by Tylan mass flow controllers. The sputter parameters maintained during the deposition of In_2O_3 films are given in Table 1. Before deposition of each film, indium target was presputtered in pure argon atmosphere for about 10 minutes to remove the surface oxide layer of the target. The films were deposited on glass substrates held at temperatures 303 K and 473 K. The temperature of the substrate was measured using copper – constantan thermocouple before the ignition of plasma for the deposition of indium oxide films. Thickness of the films was measured using multiple beam interference method. The films were characterized by studying their structure by X-ray diffraction using copper radiation with a wavelength of 0.15406 nm. The electrical resistivity and Hall measurements were carried out by using standard van der Pauw method [24]. The optical transmittance and reflectance were recorded using a Hitachi double beam spectrophotometer.

Table 1. Deposition parameters maintained during the preparation of In_2O_3 films.

Sputter target	:	Indium (100 mm dia and 3 mm thick)
Target to substrate distance	:	65 mm
Sputtering pressure (P_u)	:	4×10^{-2} mbar
Oxygen partial pressure (pO_2)	:	2×10^{-4} mbar
Substrate temperature (T_S)	:	303 K and 473 K
Cathode current density	:	1.25 mA/cm^2
Cathode voltage	:	305 V
Substrate bias voltage	:	0 to -100 V

3. Results and discussion

The deposition rate of the films was about 8 nm / min and the thickness of the films investigated was in between 80 nm and 100 nm. The ion bombardment on the substrate markedly influence the growth of the films. The substrates were kept at a known bias voltage using a dc power source. The In_2O_3 films were deposited on glass substrates held at room temperature (303 K) under various bias voltages 0 V and –100 V. Fig. 1 shows the X-ray diffraction patterns of In_2O_3 films formed at 303 K under various negative substrate bias voltages. The films formed without applying any bias were amorphous in nature. As the substrate bias voltage increased the crystallinity of the films improved. When the negative bias voltage increased the films exhibited polycrystalline nature with the presence (222) and (400) planes of the films. The intensity of the peaks increased with the increase of bias voltage indicated the improvement in the crystallinity of the films. The ion bombardment on the substrate due to negative bias voltage showed the similar effect of substrate heating. The ion bombardment increased the thermodynamics of the deposited films which is similar to the effect of substrate heating [25]. The grain size (L) of the films was evaluated from the (222) peak employing Scherrer’s formula [26]

$$L = 0.9\lambda/\beta \cos \theta \qquad (1)$$

where λ is the X-ray wavelength, β the full width at half maximum corresponding to diffraction angle θ. The grain size of the films evaluated was about 12 nm in the case of the films formed at a bias voltage of –80 V.

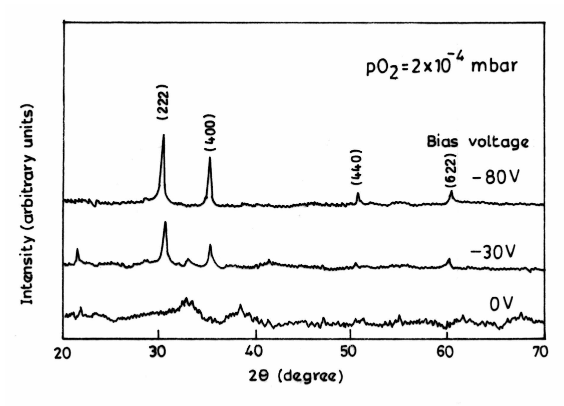

Fig. 1. X-ray diffraction patterns of In_2O_3 films formed at 303 K under various substrate bias.

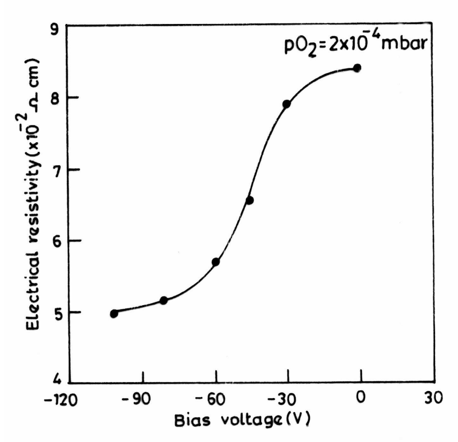

Fig. 2. Dependence of electrical resistivity of In_2O_3 films formed at 303 K on the substrate bias voltage.

Variation of electrical resistivity of In_2O_3 films formed at 303 K with the substrate bias voltage is shown in Fig. 2. The electrical resistivity of the films decreased from 8.4×10^{-2} Ω cm to 5.2×10^{-2} Ω cm with the increase of bias voltage from 0 V to –80 V thereafter it remains almost unchanged. Ion bombardment on the substrate removes preferentially oxygen atoms by resputtering resulting in the decrease of electrical resistivity. Similar decrease of electrical resistivity was also reported in sputtered indium tin oxide films [27, 28].

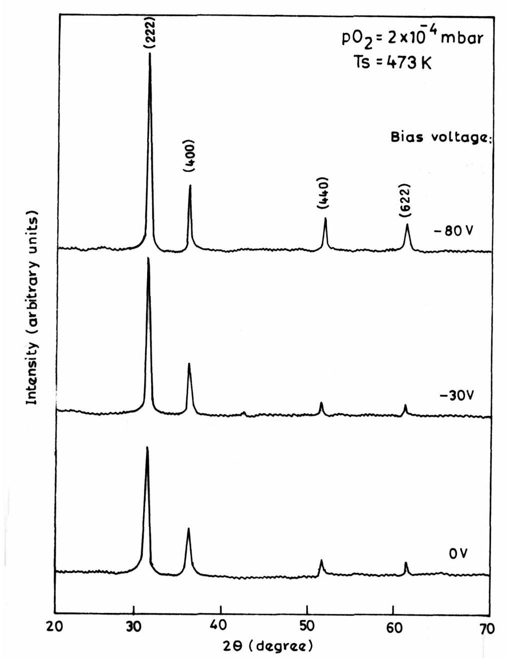

Fig. 3. X-ray diffraction patterns of In_2O_3 films formed at 473 K under various substrate bias voltage.

In order to study the influence of both the substrate bias voltage and substrate temperature, the In_2O_3 films were deposited at 473 K under various bias voltages. The quality of the films and their physical properties improved substantially by substrate heating and biasing. X-ray diffraction patterns of the films formed at 473 K under various substrate bias voltages are shown in Fig. 3. It is seen form the figure that the films formed at 473 K with out any bias voltage were polycrystalline with cubic bixbyte structure with the presence of (222), (400), (440) and (622) orientations. As the bias voltage increased the intensity of the diffraction peaks also increased. Fig. 4 shows the variation of lattice parameter and grain size with the substrate bias voltage. The lattice parameter of the films increased from 1.0080 nm 1.0122 nm with the increase of substrate bias voltage from 0 V to –80 V respectively. The lattice parameter obtained in the case of the films formed upto bias voltage of –60 V were lower than the standard value of 1.0118 nm [29]. The low value of lattice parameter at low negative bias voltages might be due to the stresses developed in the films. The grain size of the films increased from 15 nm to 23 nm with the increase of bias voltage from 0 V to –80 V respectively. The increase of grain size with the bias voltage indicated the improvement in the crystallinity of the films.

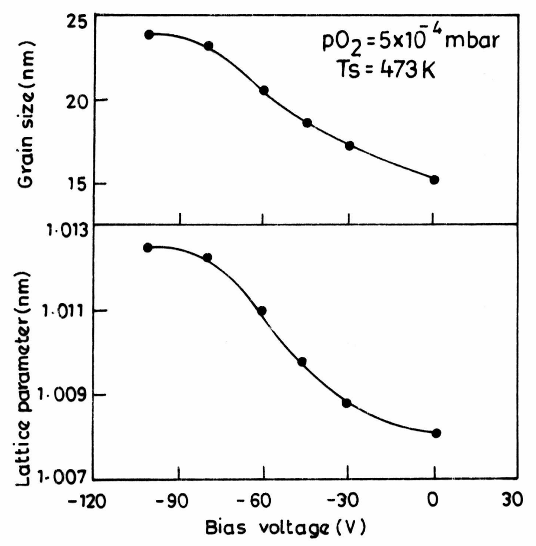

Fig. 4. Variation of lattice parameter and grain size on the substrate bias of In_2O_3 films formed at 473 K.

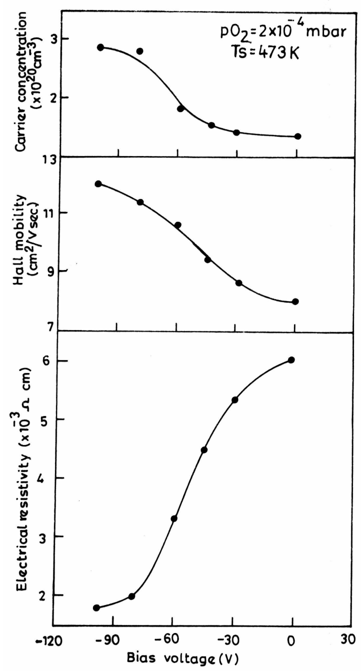

Fig. 5. Dependence of electrical resistivity, Hall mobility and carrier concentration of In_2O_3 films deposited at 473 K on the substrate bias voltage.

Fig. 5 shows the dependence of electrical resistivity, Hall mobility and carrier concentration of the films formed at 473 K on various substrate bias voltages. The electrical resistivity decreased from 6.0×10^{-3} Ωcm to 1.8×10^{-3} Ωcm, Hall mobility increased from 8 cm^2 / V. sec to 12 cm^2 / V. sec and carrier concentration increased from 1.3×10^{20} cm^{-3} to 2.9×10^{20} cm^{-3} with the increase of bias voltage form 0 V to –100 V respectively. Due to biasing and heating the substrate significantly improved the electrical properties of the deposited films. The decrease in the electrical resistivity of the films formed at 473 K with out bias voltage was due to the improvement in the alignment of grains at the grain boundaries which minimizes the trapping of the charge carriers resulting in the decrease of the grain boundary scattering. This is the phenomena generally observed in polycrystalline transparent conducting oxide films [30]. It was also observed in the case of indium tin oxide films prepared with substrate heating and biasing exhibited significant change in crystallinity and electrical properties [31,32]. The electrical resistivity achieved in the case of the films formed at 473 K and bias voltage of –100 V was 1.8×10^{-3} Ω cm, which is lower value than the reported in dc magnetron sputtered [18], reactive evaporated [7] films and comparable to that of the ion assisted deposited [16] films. The electrical resistivity of sol – gel deposited and ArF excimer laser irradiated films was 10^2 Ω cm [15]. However the films formed by pulsed laser deposition exhibited the electrical resistivity as low as 3×10^{-4} Ω cm [11]. The low electrical resistivity was also due to the increase of Hall mobility and carrier concentration.

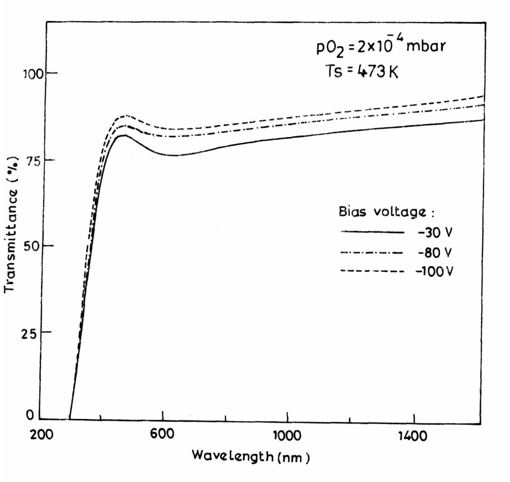

Fig. 6. Optical transmittance spectra of In_2O_3 films formed at 473 K at different substrate bias voltages.

Fig. 6 shows the optical transmittance spectra of the films formed at 473 K under various substrate bias voltages. The optical transmittance ($\lambda > 500$ nm) increased from 77% to 85 % with the increase of negative bias voltage from 0 V to –100 V respectively. The optical transmittance sharply decreased near the absorption edge in all the films. As the substrate bias voltage increased the optical absorption edge of the films shifted towards shorter wavelength side. The optical absorption coefficient (α) was evaluated from the optical transmittance (T) and reflectance (R) data using the relation

$$\alpha = (1/t) \ln [T/(1-R)^2] \quad (2)$$

where t is the thickness of the film. The dependence of absorption coefficient with the photon energy ($h\nu$) was found to obey the relation

$$(\alpha h\nu) = A (h\nu - E_g)^{1/2} \quad (3)$$

where A is the edge width parameter and E_g the optical band gap. The optical band gap of the films was evaluated by extrapolating the linear portion of $(\alpha h\nu)^2$ versus $h\nu$ at $\alpha = 0$. The optical band gap of the films increased from 3.70 eV to 3.78 eV with the increase of bias voltage from 0 V to – 100 V respectively.

Figure of merit (ϕ) is the quantity to judge the quality of the transparent conducting oxide films. The figure of merit of the films was evaluated from the optical transmittance and sheet resistance (ρ_s) using the Haacke's relation

$$\phi = T^{10} / \rho_s \quad (4)$$

The figure of merit of the films formed at 473 K increased from 1.2×10^{-4} Ω^{-1} to 1.1×10^{-3} Ω^{-1} with the increase of bias voltage from 0 V to –100 V respectively. The high value of figure of merit in the case of the films formed at the bias voltage of –100 V was due to low sheet resistance as well as high transmittance.

4. Conclusions

Indium oxide films were deposited onto glass substrates by sputtering of metallic indium target in an oxygen partial pressure of 2×10^{-4} mbar under various substrate bias voltages in the range from 0 to –100 V by employing dc reactive magnetron sputtering technique. The films formed at 303 K without substrate bias were amorphous in nature. The films deposited with substrate heating and / or substrate biasing were polycrystalline with preferred (222) orientation. The ion bombardment on the substrate during the film growth resulted in the decrease of electrical resistivity and increase of optical transparency. Indium oxide films formed at 473 K and substrate bias voltage of –100 V exhibited low electrical resistivity of 1.8×10^{-3} Ω cm, high optical transmittance of 85%, optical band gap of 3.78 eV and figure of merit of 1.1×10^{-3} Ω^{-1}.

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