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Structural, electrical and optical properties of indium–tin–oxide thin films prepared by pulsed laser deposition

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Abstract

In this work the indium–tin–oxide (ITO) thin films have been prepared by pulsed laser deposition on glass substrate. The structural, electrical and optical properties of the films have been studied as a function of substrate temperature and background deposition pressure. The dielectric function of ITO films was obtained in the wave-length range 220–2400 nm by fitting the measured transmission and reflection spectra to a dispersion relation, which combines the Drude model and Lorentz oscillator. The correlation between deposition conditions and physical properties of ITO films was observed and analysed.

Keywords: Indium–tin–oxide; Laser ablation; Optical properties; Structural properties

1. Introduction

Indium–tin–oxide (ITO) is a wide band gap semiconductor (E_g: 3.5–4.3 eV) which having low enough electrical resistivity shows the high transmission in the visible and near infrared (NIR) regions of the electromagnetic spectrum [1–7]. Moreover, the ITO has excellent substrate adherence, hardness, and chemical inertness. Because of the prominent properties this material is widely used as transparent electrodes in optoelectronic devices such as solar cells, liquid crystal displays, plasma display panels etc.

As the ITO shows the interesting and technologically important combination of properties this is probably the most widely studied transparent oxide material. However, the great majority of the most published papers is of technical nature and is limited to fabrication methods and ensuing film properties. The optical measurements are mainly used just to know how transparent the films are in the visible range of electromagnetic spectrum. The few works dealing with the calculation of dielectric function are concentrated on a particular sample [2–4] or in small (only NIR) wave length range of electromagnetic spectrum [1].

In this work amorphous, polycrystalline and highly textured ITO films were prepared on glass substrate by Pulsed Laser Deposition (PLD). The film properties were measured as a function of substrate deposition temperature (T_s) and background oxygen pressure (P_{O_2}). The dielectric function was calculated in the wide (220–2400 nm) wave length range. The correlation between structure, electrical and optical properties of the ITO films was observed and analysed.

2. Experimental details

ITO thin films were deposited on glass substrate using PLD. A Nd:YAG excimer laser (Surelite) with a wavelength of 1064 nm and pulse duration of 5–7 ns delivered an energy of 320 mJ per pulse. The laser was operated at 10 Hz and was focused through a 30 cm focal length lens onto a rotating target at a 45° angle of incidence. The energy density of the laser beam at the target surface was maintained at about 20 J/cm^2. The target-substrate distance was 40 mm. The substrate was attached with a stainless steel mask to a substrate holder, which was heated by a quartz lamp. The base pressure of the vacuum chamber before introducing oxygen was about 2×10^{-6} Torr. The target was a

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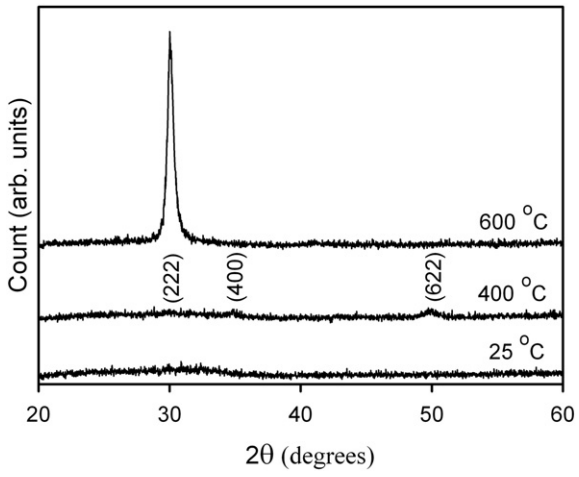

Fig. 1. The XRD patterns of ITO films grown on glass at different substrate deposition temperatures at the oxygen pressure 2.2 Pa.

commercial (Kurt Lesker Co) 90 wt.% In_2O_3 – 10 wt.% SnO_2 sintered ceramic disk of size 1×0.25 inch. No post deposition treatment was performed on all the films reported in this study.

The sheet resistance (R_s) measurements were performed using a four-probe method with help of Hewlett Packard multimeter (Hp 34401A). All sheet resistance values were determined as the average of five measurements for each film. By assuming that the thickness of the film was uniform, the film resistivity (ρ) was determined using the simple relation $\rho = R_s \times d$, where d is the film thickness.

The X-ray diffraction (XRD) data were recorded with Philips Expert Pro analytical X-ray diffractometer of (PW1710-type) using Cu Kα radiation ($\lambda = 1.54056$ Å) of 40 kV, 50 mA with the step of 0.02° and counting time 1.25 s. The optical measurements were performed by Shimadzu UV–3101 PC double beam UV–VIS–NIR scanning spectrophotometer in the wavelength range 220–2400 nm, with a slit width of 3 nm and a sampling interval of 2 nm. All the structural, electrical and optical measurements were performed at room temperature.

3. Results and discussion

3.1. Structural and electrical properties

As is seen from Fig. 1 the ITO films deposited at substrate temperature (T_s) of 25 °C are amorphous. The films deposited at 400 °C are polycrystalline. The increase of T_s up to 600 °C resulted in well crystallized and highly (222) textured films. In the studied 2θ range no peak but (222) was visible in XRD pattern of ITO films deposited at 600 °C. By fitting of the (222) diffraction peak the cubic cell parameter (a) and grain size (t) of the films were calculated. Using the found values a, the strain was calculated suggesting $a_{bulk} = 10.226$ Å [8] as a parameter of unstrained cell. The results are summarized in the Table 1.

As seen from the Table 1 the ITO films are tensionally strained. One of the possible reason of the strain is the difference in the thermal expansion coefficients of ITO ($\alpha_f = 10.2 \times 10^{-6}$ K^{-1} [9]) and glass substrate ($\alpha_s = 4.6 \times 10^{-6}$ K^{-1}). At cooling after film deposition the glass substrate shrinks up much lower then ITO films that is the reason of the tension strain in films. The estimated values of strain $\varepsilon = (\alpha_f - \alpha_s)\Delta T \approx 2.1 \times 10^{-3}$ for films deposited at 400 °C and $\varepsilon \approx 3.2 \times 10^{-3}$ in the case of $T_s = 600$ °C are of the same order of values found from XRD.

The average grain size (t) was defined follow the Scherrer equation:

$$t = \frac{K\lambda}{B\cos(\theta)}, \quad (1)$$

where $K = 0.9$ is a crystal shape constant, λ is the X-ray wavelength, B is the width (in radians) of a reflection measured at half of its height and θ is the Bragg diffraction angle. The calculations revealed that as T_s increases from 400 to 600 °C, the grain size of the films increases from ≈10 up to ≈29 nm (see Table 1). In the case of films deposited at 600 °C we observed the strong increase (up to 91 nm) of t as P_{O_2} decreases down to $P_{O_2} = 0.7$ Pa. The further decrease of P_{O_2} resulted in the decrease of t.

The increase of the grain size with T_s is obviously caused by the higher migration possibilities of deposited particles within the substrate surface at higher temperatures. The observed influence of oxygen pressure on crystallinity the most probably is linked with the energy of bombardment particles during the film growth. Collisions of the ablated particles with gas molecules decrease under lower P_{O_2}. So, the energy of particles reached the substrate is higher that helps their migration on substrate surface, better arrangement of crystalline structure and resulted in lower strain and bigger grain size with decreasing oxygen pressure.

As is seen from the Table 1 the electrical properties of the grown ITO films also show the strong dependence on P_{O_2}. The decrease of ρ with P_{O_2} can be explained by the fact that the

Table 1
The results of structural and electrical study

Sample	Growth temperature (°C)	Oxygen pressure (Pa)	Lattice parameter (Å)	Strain (10^{-3})	Grain size (nm)	Sheet resistance (Ω/sq.)	Resistivity (10^{-5} Ω m)
1	25	2.2				122.3	1.6
2	400	2.2	10.297	6.9	10	64.20	0.88
3	600	2.2	10.316	8.8	29	51.99	0.73
4	600	1.5	10.281	5.4	59	23.94	0.36
5	600	1.0	10.269	4.2	82	20.39	0.30
6	600	0.7	10.263	3.6	91	16.29	0.24
7	600	0.5	10.265	3.8	75	15.30	0.22