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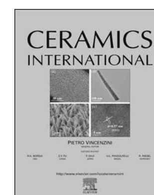

Highly conducting and transparent antimony doped tin oxide thin films: the role of sputtering power density

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ABSTRACT

Sb doped SnO_2 thin films were deposited on quartz substrates by magnetron sputtering at 600 °C and the effects of sputtering power density on the preferential orientation, structural, surface morphological, optical and electrical properties had been studied. The XRD analyses confirm the formation of cassiterite tetragonal structure and the presence of preferential orientation in (2 1 1) direction for tin oxygen thin films. The dislocation density analyses reveal that the generated defects can be suppressed by the appropriate sputtering power density in the SnO_2 lattice. The studies of surface morphologies show that grain sizes and surface roughness are remarkably affected by the sputtering power density. The resistivity of Sb doped SnO_2 thin films gradually decreases as increasing the sputtering power density, reaches a minimum value of 8.23×10^{-4} Ω cm at 7.65/cm^2 and starts increasing thereafter. The possible mechanisms for the change in resistivity are proposed. The average transmittances are more than 83% in the visible region (380–780 nm) for all the thin films, the optical band gaps are above 4.1 eV. And the mechanisms of the variation of optical properties at different sputtering power densities are addressed.

1. Introduction

Transparent conductive oxide (TCO) thin films have been investigated for their potential use in light-weight, flat panel applications, including cell phones, liquid crystal displays (LCD), electronic paper, touch screens, and solar cells due to their excellent optical and electrical properties [1–5]. Currently, tin doped indium oxide (ITO) thin films are widely used as TCOs because of their low resistivity in the range of (10^{-4} Ω cm), high light transmittance (≥80%) in the visible region and ease of wide-area deposition by simple techniques [6–8]. However, indium is scarce in the nature, and is estimated to be 0.05 ppm on earth, the price of ITO is expensive and increasing due to the high demand in a large variety of applications [9,10]. Alternative transparent conducting thin films are therefore of great technological and scientific interest.

Tin oxide (SnO_2) is a II-IV semiconductor with a wide and direct band gap of 3.6 eV [11], it is chemically stable in acidic and basic solutions, thermally stable in oxidizing environments at high temperature, and also mechanically hard [12,13]. These advantages make it a promising alternative to ITO. The electrical properties of tin oxide have been further improved by incorporation of numerous doping elements such as F [12,14], Sb [15,16], As [17], Cd [18] and W [19]. Among these, Sb doped SnO_2 (ATO) thin films in particular constitute a promising candidate as they exhibit quasi metallic conductivity maintaining at the same time their transparency in the visible range [15]. Several technologies have been proposed for the preparation of ATO thin films, including magnetron sputtering [20,21], thermal decomposition [22], plasma-enhanced chemical vapor deposition [23], sol-gel [24], plasma-assisted molecular beam epitaxy [25], pulsed-laser deposition [26], spray pyrolysis [27], et al. Among these techniques, rf-magnetron sputtering is one of the most appropriate deposition techniques for high quality oxide films, and gives better adhesion, larger coverage and higher film density than other methods [28,29]. For the magnetron sputtering deposition of thin films with regards to the crystal structure, the crystal quality strongly depends on the sputtering conditions, such as sputtering power density, deposition time, substrate-target distance, substrate temperature, ambient gas, sputtering pressure, the nature of substrate and target constitution [30,31]. The deposition process is affected by the bombardment of the growing film with species from the sputtering target and the plasma [32]. In addition, the sputtered atoms with energies in the range up to 20 eV [33], higher energy ions from the plasma and neutral atoms reflected at the target hit the growing film. Therefore, studies on understanding the effect of sputtering power density on the structure,

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surface, optical and electrical properties are very important.

In the present work, high transparent and low resistive Sb doped SnO_2 (ATO) thin films were prepared on quartz substrates by radio frequency (r. f.) magnetron sputtering. A high transmittance of ~84% and a low resistivity of 8.83×10^{-4} Ω cm were achieved. The dependence of structure, preferential orientation, surface morphological, optical and electrical properties of ATO thin films on sputtering power density has been investigated.

2. Experimental

Sb doped SnO_2 (ATO) thin films were prepared on quartz glass substrates by using radio frequency (r. f.) magnetron sputtering system. The ATO target was prepared by the SnO_2 powder (purity 99.9%) and Sb_2O_3 powder (purity 99.5%), and the mol proportion was 88: 6. These two kinds of powders were mixed in an agate mortar for 3 h. After pre-sintered at 1250 °C for 6 h, the powders were pressed into 2-in. diameter and 5 mm thickness pellet at 15 t and then sintered at 1450 °C for 6 h in air. The substrates were cleaned by isopropyl alcohol, acetone and deionized water for 20 min, respectively. The target to substrate distance was maintained as 60 mm. Before sputtering, the vacuum chamber was evacuated down to a base pressure of 6.0×10^{-6} Torr by using a molecular pump. The substrate temperature was set as 600 °C. High purity (99.99%) O_2 and (99.99%) Ar were introduced through separate mass flow controllers with the ratio of 1: 20, and the total pressure was maintained at 8 mTorr. Pre-sputtering was applied for 10 min to remove any contaminants on the surface of ATO target. The sputtering power densities were set as 2.55 W/cm^2, 5.10 W/cm^2, 7.65 W/cm^2 and 10.20 W/cm^2, respectively. The thickness of the ATO thin films was controlled at 300 nm by controlling the deposition rate and time.

The X-ray diffraction (XRD) analyses were carried out by X-ray diffractometer (Rigaku D/MAX–RB, Akishima, Tokyo, Japan) in the range of 20–70°. Surface morphologies were characterized by the atomic force microscopy (AFM) using a Nanoscope Multimode 8 (Bruker, Santa Barbara, CA, USA). The thickness was measured by Alpha-Step D-100 profilometer (KLA-Tencor, California, USA). Room temperature Hall effect measurements were carried out using a Hall measurement in the van der Pauw configuration (Ecopia HMS 3000 Hall System, Republic of Korea). The sheet resistance was measured by four-point probe instrument (SX1934, SuZhou, PR China). The transmittance was measured using an ultraviolet-visible (UV–Vis) spectrophotometer (Varian Cary 5000) in the wavelength range 200–800 nm.

3. Results and discussions

The X-ray diffraction analysis was carried out by recording the X-ray diffractometer patterns in the range of 20–70° to investigate the crystal structure and preferred crystal orientation of the ATO thin films. Fig. 1 shows the XRD spectra of ATO thin films prepared at 2.55, 5.10, 7.65 and 10.20 W/cm^2. It can be seen that the main growth directions are (1 1 0), (1 0 1), (2 0 0), (2 1 1), (2 2 0), (2 2 1) and (3 0 1), which reveal that all the Sb doped SnO_2 thin films are polycrystalline and retain the cassiterite tetragonal (rutile type) structure [34,35]. No characteristic peaks of Sn, Sb_2O_3 or Sb_2O_5 can be observed in the XRD patterns, which means that antimony atoms substitutionally replace tin in the tetragonal lattice or antimony atoms segregate into the non-crystalline region in grain boundary. As shown in Fig. 1, the peak intensity of the ATO thin films obviously increased with increasing the sputtering power up to 7.65 W/cm^2 and then slightly decreases with further increase of sputtering power density, but the locations of the diffraction peaks do not change significantly. The change of peak intensity is related to the change of crystallinity, the better the crystal quality of thin films is, the more intense the peak intensity becomes. That is to say, the ATO thin films deposited at 7.65 W/cm^2 have the best crystal quality.

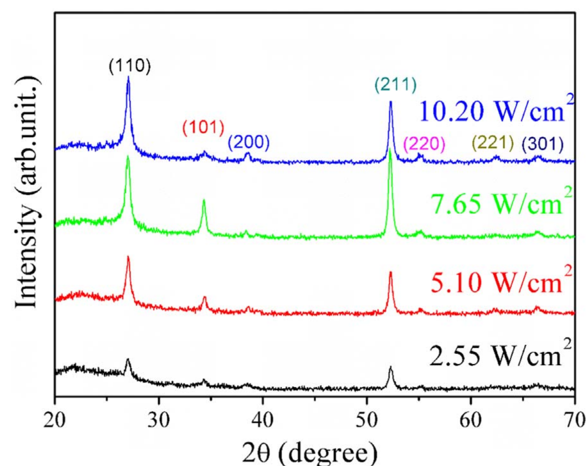

Fig. 1. XRD spectra of ATO thin films prepared at sputtering power densities of 2.55, 5.10, 7.65 and 10.20 W/cm^2.

The preferential orientation of thin films with polycrystalline nature can be understood from the texture coefficient values for all planes. In general, the texture affects the structural, electrical and optical properties of the thin films as well as the performance and reliability of the fabricated devices. The texture analysis was quantitatively carried out from the $K\alpha_1$ component of each diffraction peak in the framework of the Harris method [36], and the texture coefficients $P(h_i, k_i, l_i)$ for each (h_i, k_i, l_i) crystallographic direction were respectively defined as follows [36,37]:

$$P(h_ik_il_i) = \frac{I(h_ik_il_i)}{I_0(h_ik_il_i)}\left[\frac{1}{n}\sum_{i=1}^{n}\frac{I(h_ik_il_i)}{I_0(h_ik_il_i)}\right]^{-1} \quad (1)$$

where $P(h_i, k_i, l_i)$ is the texture coefficient for each (h_i, k_i, l_i) crystallographic direction, I is the observed intensity of (h_i, k_i, l_i) plane, I_O is the standard intensity, and n is the number of reflections observed in the XRD pattern. Fig. 2 shows the texture coefficients for each crystallographic direction as a function of sputtering power density. The relative intensity between the different (h_i, k_i, l_i) Bragg reflections is strongly dependent upon the sputtering power density. According to analyzing the texture coefficient values of all planes, the preferential orientation of polycrystalline thin films can be understood. For the (2 1 1) plane of the films, $P(h_i, k_i, l_i) > 1$, which indicates that the crystallite films are preferentially oriented along the (2 1 1) plane. The $P(h_i, k_i, l_i)$ of (2 1 1) plane initially increases with sputtering power density, reaches a maximum at 7.65 W/cm^2, and decreases as the sputtering

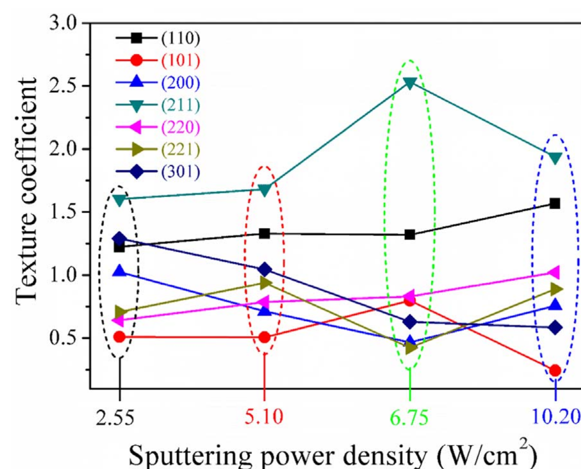

Fig. 2. Texture coefficients for each crystallographic direction of ATO thin films prepared at sputtering power densities of 2.55, 5.10, 7.65 and 10.20 W/cm^2.

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