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Control of preferred (222) crystalline orientation of sputtered indium tin oxide thin films

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ABSTRACT

We report a two-step growth process for the fabrication of (222)-plane textured indium tin oxide (ITO) films. A thin ITO seed layer was grown in mixed Argon + Oxygen gases, followed by a thick ITO deposited in Argon gas. X-Ray diffraction shows that the sputtered ITO films exhibit strongly preferred (222) crystalline orientation. The (222)-plane textured ITO films have high transmittance above 80% in the visible range and carrier concentration, mobility and resistivity in the range of 10^{21} cm^{-3}, 40 cm^2/Vs and 10^{-4} $\Omega\cdot$cm, respectively. The surface roughness of our (222) textured ITO films is 1.4 nm, which is one of the smallest value obtained from sputtered ITO thin films.

1. Introduction

Tin-doped indium oxide (ITO) has been known as a transparent electrode in several optoelectronic devices such as liquid crystal displays, solar cells, organic light-emitting diodes (OLEDs), smart window, touch screen, and other flat panel displays due to its high optical transmittance and low electrical resistivity [1–6]. Recently, the organic light-emitting diodes (OLEDs), which are one of the most promising candidates for flat panel displays, demand a very flat surface of ITO film [7] for improving electroluminescence efficiency and display lifetime [8]. In general, homogeneity and surface roughness are very important for the reliability of devices since the organic layers in the OLEDs have thicknesses of only about 100 nm [9]. In particular, the peak-to-valley roughness of ITO film has a linear relationship with the reverse leakage current of devices [8]. Also, the surface morphology of ITO films considerably affects the patterning properties during the fabrication process of flat panel displays [10]. Many published literatures show that electrical and optical properties of ITO thin films strongly depend on its preferential crystallographic orientation. The ITO films with (400) crystallographic orientation have smaller optical band-gap, less effective "Sn" doping and larger grain size than the (222) textured films [11]. Nakaya et al. proposed that the ITO films with the (222) preferred orientation experience little deterioration at its interface with an over-lying film, thereby improving the light emission characteristics and lifetime of devices [12]. In addition, since there is a small lattice mismatch between the neighboring oxygen-oxygen (O-O) distance on the close-packed ITO (222) and ZnO (002) planes, it benefits the initial nucleation and subsequent growth of high quality ZnO materials on (222) ITO substrates [13,14], probably leading to a good contact for carrier transport in solar cells based on ZnO substance materials.

Kim et al. have found that the preferential orientations of the ITO thin films depend on the oxygen partial pressure. An ITO film grown with pure Ar gas shows a preferential (400)-plane orientation parallel to the substrate surface while the preferential orientation of films changed from (400) to (222) plane when even a small amount of O_2 was added to the Ar sputtering environment. It was also observed that the diffraction intensity of the (222) peak decreased as the oxygen partial pressure increased [15]. Moreover, most publications indicate that the (222) textured ITO films grown in mixed Ar + O_2 gases have poor conductivity compared with the (400) textured ITO films grown in Ar gas environment. The reason for this is the reduction density of oxygen vacancies, which is the main contributor of electrical carriers in the ITO film.

In this paper, we report the procedure to prepare (222) textured ITO films with high conductivity grown in Ar gas environment instead of mixed Ar + O_2 gases. The proposed procedure is the two-step sputtering process, in which a thin oxygen seed layer of indium tin

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oxide (O-ITO) was sputtered in the mixed Ar + O_2 gases prior to the deposition of the thick overhead ITO films in Ar gas.

2. Experiments

The ITO thin films were prepared on soda-lime glass substrate by dc magnetron sputtering. The target was commercial ceramic target with 10 wt.% SnO_2 (99.99% purity) impurity. The substrate was kept at a distance of 5 cm from the target. The substrate temperature and sputtering power were maintained at 350 °C and 50 W during the deposition, respectively. In order to deposit an oxygen seed layer of indium tin oxide (O-ITO), the vacuum chamber was evacuated down to pressure 5.3×10^{-4} Pa prior to deposition. Then the oxygen reactive gas was introduced into the chamber and the required pressure, for example 4.2×10^{-1} Pa, was set. Argon gas was introduced thereafter till the preset pressure reached 5.3×10^{-1} Pa. Both argon inert gas flow and oxygen reactive gas flow were controlled by a mass flow controller. The thin O-ITO seed layer was firstly grown on glass in mixed (O_2 + Ar) gases at 5.3×10^{-1} Pa. The thickness of O-ITO seed layer is about 2 nm. Then, the vacuum chamber was evacuated down to pressure 5.3×10^{-4} Pa again for the following deposition of the 300-nm thick ITO layer in pure Ar gas at 5.3×10^{-1} Pa. The thickness of films was monitored by using the Quartz oscillator (XTM/2-INFICON (USA)). The crystalline phases of the films were characterized in the θ–2θ mode by using a D8 Advance (Bruker) X-ray diffractometer (XRD) with Cu Kα radiation ($\lambda = 0.154$ nm). Electrical properties of films were carried out using Hall measurements (Ecopia HMS-3000). The optical transmittance spectra were measured using a UV–vis (Jasco V-530) in the wavelength range from 200 nm to 1100 nm. The surface morphology was investigated by Atomic force microscopy (5500 AFM SYSTEM-AGILENT, Tapping mode) and scanning electron microscopy (SEM, JEOL JSM-7401F, operating voltage is 30 kV). The work function was measured by Ultraviolet Photoelectron Spectroscopy (UPS) using a Model AC-2 instrument (RIKEN KEIKI).

3. Results and discussion

Fig. 1 shows X-ray diffraction patterns of the ITO thin films prepared with and without the O-ITO seed layer. It is obvious that the ITO thin film without O-ITO seed layer reveals polycrystalline structure with differently orientated crystalline planes such as (400), (222), (211), (440), and (622). Among these planes, it has been found that there is preferential growing competition between (222) and (400) planes, the (400) plane preferential orientation. This structural characteristic has been attained by other authors by growing ITO thin films in pure Ar gas and using not only magnetron sputtering but also other methods [16–19]. In contrast, the ITO film with O-ITO seed layer, shows a prominently strong (222) peak, which can be understood that grain growth in the (222) direction is obviously favored against growth in other directions [20]. This indicates that the thin O-ITO seed layer has significant effect on the crystal grain orientation of an overhead ITO film.

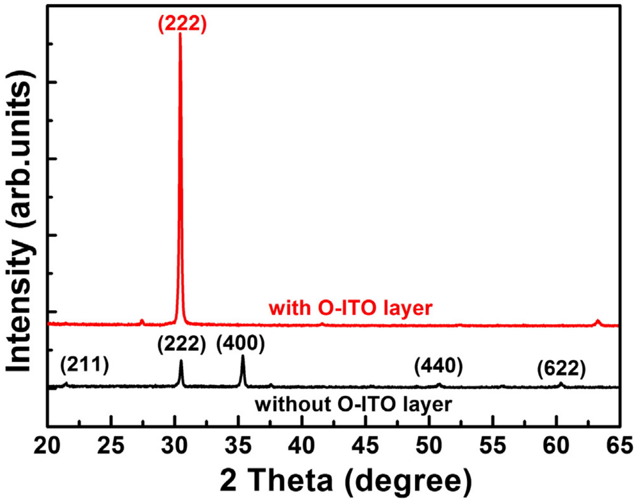

Fig. 1. X-ray diffraction patterns of ITO thin films with and without O-ITO layer.

In addition, SEM images reveal the significant influence of the O-ITO seed layer on surface morphology of the overhead ITO layer. The visible difference of surface morphology of the ITO thin films prepared with and without the O-ITO seed layer is shown in Fig. 2. It can be seen that the ITO film with O-ITO layer reveals “grain structure” (Fig. 2a) while the film without O-ITO layer shows “domain structure” (Fig. 2b). This “domain structure” is also called a “grain–subgrain” structure [10] or “domain–grain” structure [7] of conventionally sputtered ITO thin films in an oxygen-deficient environment, which has been obtained by other authors [7,10,21]. The SEM images strongly show that there is transformation from “domain structure” into “grain structure” corresponding to the (400) into (222) texture, respectively, due to an introduction of the initial O-ITO seed layer prior to conventionally deposited ITO layer only in pure Ar gas.

Fig. 3 exhibits the estimated surface roughness (RMS) obtained from AFM analysis. There are distinct differences in surface roughness between the two samples with and without O-ITO layer of 1.4 nm and 3.7 nm in a scan area of 5 μm × 5 μm, respectively. Jung et al. [7] reported that the ITO samples prepared by dc magnetron sputtering have an RMS roughness in the range 2–4 nm. Raoufi et al. showed AFM images of as-deposited and annealed ITO thin films revealing the formation of a porous granular surface with surface roughness values in the range of 0.847–3.846 nm [22]. Hotovy et al. reported that ITO thin films grown

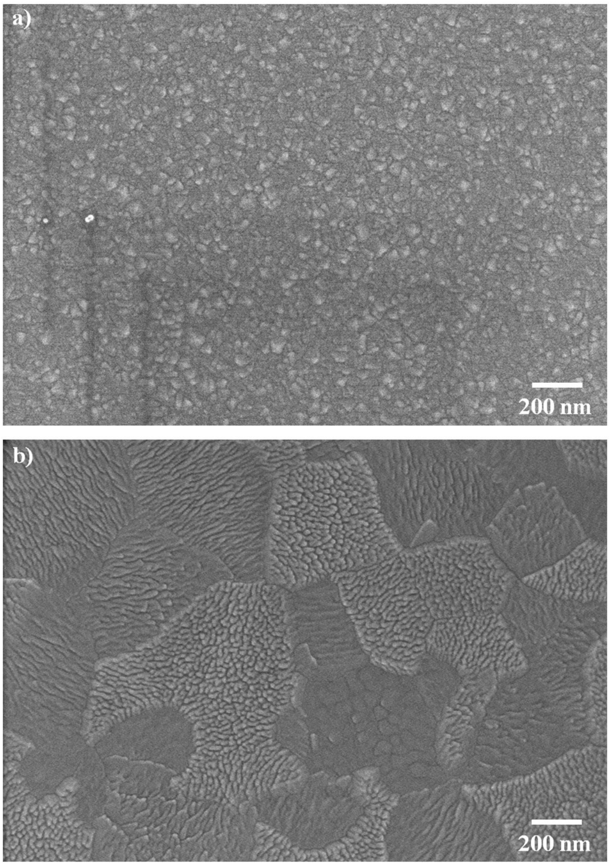

Fig. 2. SEM images of a) the (222) textured ITO film with O-ITO layer and b) the ITO without initial O-ITO layer.