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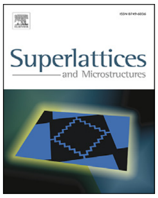

Improved conductivity of indium-tin-oxide film through the introduction of intermediate layer

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ABSTRACT

A thin intermediate layer (Ag, AuSn, In, Ni, Sn, SiO_2) was individually deposited on glass substrates prior to the deposition of indium-tin-oxide (ITO) thin film by radio-frequency (RF) magnetron sputtering employing ITO target (composition ratio of In_2O_3:SnO_2 = 9:1). The structural, optical and electrical properties were investigated to compare the ITO thin film with and without an intermediate layer. The preferential orientation of all ITO films was along (222) plane. Although all thin films were polycrystalline, the presence of intermediate layer promoted the overall crystallinity. The sheet resistance and resistivity of the ITO film were reduced from ~68 Ω/□ to ~29–45 Ω/□, and 16.2×10^{-4} Ω cm up to 7.58×10^{-4} Ω cm, respectively, by inserting a thin metal layer underneath the ITO film, and it is dependent on the degree of crystallization. The optical transmittance in the visible region varies from 40 to 88% for different samples. Based on the evaluation from Tauc plot, the optical band gap falls in the range of 4.02–4.12 eV. Physical film thickness was compared with that evaluated by optical measurement in the visible range and the physical thickness was found to be smaller. Similarly, the carrier concentration/scattering time from Hall effect measurement were also compared with that from optical measurement in the infrared region. Haacke's figure of merit (FOM) was employed to assess the quality of the ITO films, and the highest FOM is credited to ITO/In up to ~8×10^{-3} Ω^{-1} in the visible light region.

1. Introduction

Indium-tin-oxide, Sn:In_2O_3 or more commonly known as ITO, has wide applications as transparent electrode for liquid crystal displays, flat panel displays, plasma displays and electronic ink applications. On top of its vast usages in display technologies, ITO emerges as a key component in organic light emitting diodes (OLED), dye-sensitized solar cells (DSSCs), fiber optic devices, antistatic coatings and aircraft windshield [1,2].

Numerous research have been carried out on fluorine-doped tin oxide (FTO), aluminum-doped zinc oxide (AZO), indium-doped zinc oxide (IZO), indium-gallium-doped zinc oxide (IGZO), indium-tin-doped zinc oxide (ITZO) to seek for alternative transparent conducting oxide (TCO) [3–5], presently, ITO remains as the most promising TCO material [6]. For any TCO, the film emphasizes on both high electrical conductivity and high optical transparency. Therefore, the challenge to prepare an

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ideal ITO film lies in the improvement of the film from both aspects, but often the enhancement in electrical conduction is accompanied by the degradation of its optical transparency, or vice versa.

The ITO films sputtered at room temperature are normally amorphous or less crystallized, resulting in high electrical resistivity. Therefore, various post-deposition treatments are desired to improve the film crystallinity. These include electro-annealing ITO film by using electric current to generate heat, corresponds to temperatures 100–300 °C [2,7], and surface modification of ITO film by plasma treatment in Ar, N_2, O_2 or H_2 ambient [8–10]. A simpler path is through thermal annealing of the as-deposited films at different temperatures 100–600 °C [11,12]. Alternatively, substrate temperature ranging 100–450 °C have been applied during the sputtering process to replace the post deposition calcination [13]. Although these attempts have enhanced the conductivity, further improvement is desirable for advanced applications.

Recent approaches insert a metal layer between two ITO films to form a multilayer structure, ITO/metal/ITO, or even sandwich the ITO film by two metal films, metal/ITO/metal. Despite metal/ITO/metal structure offers very low sheet resistance, it suffers a huge reduction in optical transmittance due to two opaque metal layers [5,11]. ITO/metal/ITO in contrast, is more popular because this combination possesses lower sheet resistance than a single ITO film of similar thickness [14].

To date, the frequently investigated metal layers for ITO/metal/ITO structure focus on the precious metals Pt, Ag, Au, and a few studies are based on Cu and Ni [6,14,15]. These materials are favored due to their low resistivity, in particular, Ag and Cu of lowest resistivity among all bulk metals [16]. For such multilayer structure, various thickness combinations for the top and bottom ITO layer have been examined. Besides, metal layer thickness ranging from 5 to 35 nm have been sandwiched between two ITO films [16–18]. To name a few, ITO/Cu/ITO and ITO/Ni/ITO achieved a maximum figure of merit (FOM) of 5.2×10^{-4} and $2 \times 10^{-3}\ \Omega^{-1}$, respectively. Better FOM was seen in ITO/Au/ITO up to $6.6 \times 10^{-3}\ \Omega^{-1}$ and ITO/Ag/ITO up to $26.1 \times 10^{-3}\ \Omega^{-1}$. These studies concluded that the thickness of each layer significantly influenced the resulting electrical conductivity and optical transmittance [17–20].

A less complicated structure with the introduction of single metal layer to an ITO film i.e. ITO/metal and metal/ITO, is deemed possible to enhance the properties of the ITO film. In comparison to the tri-layer structure discussed earlier, only a few works are reported based on the bilayer combination which requires less control during the fabrication process. Ni/ITO, ITO/Ag and ITO/Ni deposited on glass and silicon substrates were post thermal annealed in 450–650 °C to improve the film conductivity and transmittance [21,22]. In lieu of a compact metal thin film, Jeong et al. implemented Ag grid lines of different girds spacing beneath ITO film and accomplished an average FOM of $100 \times 10^{-3}\ \Omega^{-1}$ [23].

In this study, in addition to Ag and Ni, we explore the possibility to employ other intermediate layers, including AuSn alloy, In, Sn and SiO_2 underneath ITO film and investigate the effects of these layers toward the bilayer structure. A comparison between ITO and ITO/intermediate-layer films was carried out for their structural, optical and electrical properties as well as the relationship among the mentioned properties.

2. Experimental

Glass substrates were cleaned by ultrasonication in alkaline solution, followed by several rinsing in deionized water to remove organic contaminants and native oxides. A variety of intermediate layers including silver (Ag), gold80-tin20 alloy (AuSn), indium (In), nickel (Ni), tin (Sn) and silicon dioxide (SiO_2) were separately evaporated onto the glass substrates by using a thermal evaporator unit (Edwards AUTO 306). SiO_2 layer was obtained through the evaporation of silicon (Si) and post thermal treatment at 500 °C for 1 h to achieve highly transparent SiO_2 layer. Whereas all metals (above 99.9% purity) was directly deposited on glass substrates and did not undergo any post treatment. Evaporation of all materials was performed at chamber pressure 7.0×10^{-5} mbar at room temperature.

An ITO target with In_2O_3:SnO_2 composition ratio 9:1 was employed to sputter the ITO layer by RF magnetron sputtering unit (Auto HHV 500) on the pre-deposited intermediate layer. The chamber base pressure was initially reduced to 3.3×10^{-5} mbar and the substrate temperature was set to 300 °C in a pure argon ambient. The RF power was fixed at 100 W and the working pressure was maintained in the range of $5–7 \times 10^{-3}$ mbar. A total deposition time of 45 min with deposition rate saturated at 0.9–1.0 Å/s was required to achieve a film thickness of approximately 250 nm. The bare ITO film on glass substrate is referred as ITO, whereas the remaining samples are denoted as ITO/x, where x is the intermediate layer deposited on glass substrates.

For materials characterization, the structural properties were measured by high-resolution X-ray diffractometer (HR-XRD) (PANalytical X'pert PRO MRD PW3040) with CuKα_1 source of 0.154 nm wavelength. The elemental composition of the samples was confirmed by energy dispersive X-ray spectroscopy (EDX) whereas the physical thickness of the thin film was imaged by field-emission scanning electron microscope (FE-SEM) (Nova NanoSem 450). Electrical properties were evaluated by Hall effect measurement system (Lakeshore controller 601/DRC-93CA). Optical transmittance were recorded by ultraviolet–visible–near infrared (UV–Vis–NIR) spectrophotometer (Agilent Cary 5000).

3. Results and discussion

The ITO/x films were investigated from different aspects including structural, electrical and optical analysis, as well as the relation among these properties.