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Effects of Tin Concentration on the Electrical Properties of Room-Temperature Ion-Beam-Assisted-Evaporation-Deposited Indium Oxide Thin Films

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Tin-doped indium oxide (ITO) thin films were deposited at room temperature by a dual-oxygen-ion-beam-assisted evaporator system and the effects of doped tin concentrations in the films on the electrical properties of the ITO films were investigated. Doped tin atoms in amorphous ITO films caused extra scattering and structural defects due to the inactivation of tin atoms in the films. Therefore, increasing the tin concentration decreased the conductivity. The lowest resistivity of indium oxide (IO) and ITO obtained was 3.6×10^{-4} Ω-cm for IO and 4×10^{-4}–7×10^{-4} Ω-cm for 5–25 wt% SnO_2. The mobility and mean free path of these films were 20–63 cm^2/V·s and 3.3–7.4 nm, respectively. [DOI: 10.1143/JJAP.41.L999]

KEYWORDS: ITO, room temperature, amorphous, scattering, defect

Recently, tin-doped indium oxide (ITO) thin film deposited at room temperature (RT) or low temperature (below 100°C) has been proposed for use with flexible plastic substrates such as polycarbonate, polyethylene terephthalate, and polyimide for the next generation of flat panel display devices. In general, ITO films deposited below 150°C have characteristic amorphous crystal structures.[1–4] Amorphous ITO films (a-ITO) can also be beneficial to current flat panel display devices because amorphous films decrease the production cost of patterned electrodes due to having a much higher etching rate than crystalline ITO films and showing clearer lithographic patterning.[3,5]

RT-ITO films have been deposited by several techniques such as evaporation,[6–8] high-density-plasma-assisted evaporation (HDPE),[9–11] ion-beam-assisted evaporation (IBAE),[12–17] magnetron sputtering,[1–3] ion beam sputtering (IBS),[18–21] and pulsed laser deposition (PLD).[4,22,23] Among these techniques, sputtering has been widely used as a large-area deposition technique. However, ITO film deposited by the sputtering tends to show two different microstructures in the film, i.e., the top layer is composed of a polycrystalline structure while the bottom layer is amorphous.[24] These different structures have unfavorable effects on electrical, optical, and etching properties for opto-electronic applications.[3] On the other hand, films deposited by evaporation maintain an entirely amorphous structure below 150°C without showing a polycrystalline structure.

Electrical properties of amorphous IO (a-IO) and ITO films depend strongly on oxygen vacancies and the concentration of tin atoms. The former donate two free electrons per vacancy to the donor level through ionization while the latter decreases the conductivity as an extra scattering center.[4,9,20,25,26]

In this study, the role of tin concentration in a-ITO thin films on electrical properties such as resistivity, mobility, and mean free path has been investigated for the films deposited by the IBAE technique at RT while varying oxygen concentration under optimal conditions.

Amorphous ITO thin films with five different tin concentrations were deposited on soda-lime glasses by a dual oxygen ion-beam-assisted evaporation system. The source materials were sintered ITO bulks which contain 0, 5, 10, 15, and 25 wt% tin oxide, respectively. The deposition system used in the experiment consisted of an electron beam evaporator and two internally mounted oxygen ion guns. Both of the oxygen ion guns were driven by rf inductively-coupled plasma (13.56 MHz) and two grids were attached for the extraction (−100 V) and acceleration (+1.2 kV) of the oxygen ions generated. To study the effect of the flux of the oxygen ion beam irradiated to the substrate, one of the oxygen ion guns (ion gun 2) was set facing the substrate and the flux was varied by changing the rf power (200 to 400 W) and oxygen flow rate (4 to 6 sccm). The other oxygen ion gun (ion gun 1) was located near the ITO evaporator and irradiated an oxygen ion beam towards the ITO flux, not towards the substrate. The flow rate of ion gun 1 was kept constant at 4 sccm. The details of the system and its operational conditions can be found elsewhere.[14,15] The properties of the deposited a-ITO films were measured using a four-point probe and a Hall measurement system.

Figure 1 shows the effect of SnO_2 wt% in the ITO evaporation source on the resistivities of the deposited ITO films measured by the Hall measurement system. The carrier concentrations of the films containing 0 to 25 wt% tin oxide were varied by controlling the oxygen ion beam flux to the sub-

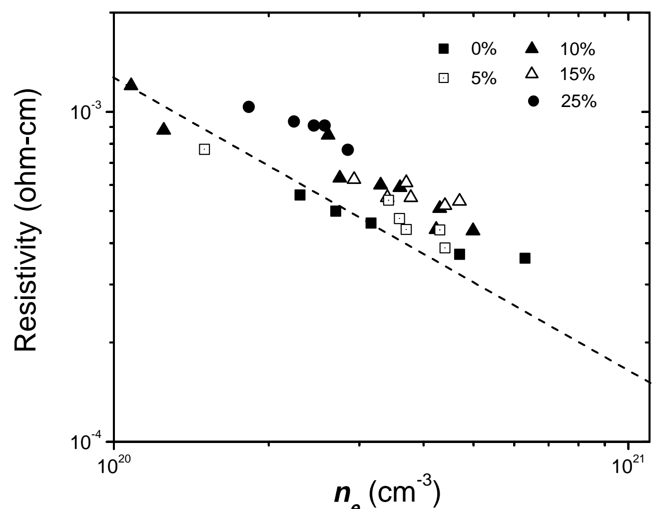

Fig. 1. Resistivity of amorphous IO and ITO thin films as a function of free electron concentration for various tin oxide weight percentages, 0 wt%, 5 wt%, 10 wt%, 15 wt% and 25 wt%. The dotted line was calculated by Dingle using the Born approximation.[28]

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strate (oxygen flow rate to ion gun 2) during the deposition. The data was reconfigured to compare with a theoretical relationship between the resistivity of ITO and carrier concentration suggested by Bellingham *et al.*[9,27] and Dingle[28] using the Born scattering approximation for ionized impurity scattering (shown as a dotted line). As shown in the figure, the experimental data measured in this experiment appear to follow the relationship for pure indium oxide, and are closer to the dotted line at the lower tin concentration in the ITO film. The increased deviation from Dingle's theoretical value with the increase of the tin doping is ascribed to the increase of neutral impurity scattering by inactivated tin atoms in the film. The insensitivity of electrical resistivity to the electron concentration at the high electron concentration regime appears to be related to the increased possibility of multiple scattering as suggested by Bellingham *et al.*[9] In our experiment, the carrier concentration ranged from $3 \times 10^{20}/\mathrm{cm}^3$ to $5 \times 10^{20}\ \mathrm{cm}^{-3}$.

The relationship between the carrier concentration and the carrier mobility for different tin concentrations is shown in Fig. 2 to investigate the effect of tin percentage in the ITO film on the carrier mobility. As shown in the figure, the increase of carrier and tin concentration decreased the carrier mobility. The decrease of carrier mobility may originate from various scattering mechanisms. In the case of ITO, the scattering can be classified as 1) ionized impurity scattering, 2) neutral impurity scattering, 3) grain boundary scattering, and 4) lattice scattering. Among these, lattice scattering (4) is not the major scattering mechanism at low temperatures.[29–31] Also, because the grain size of room-temperature-deposited ITO is known to be above 10 nm,[28] if the mean free path (MFP) of the electron in ITO is less than 10 nm, the grain boundary scattering (3) can also be ignored.

Figure 3 shows the MFP calculated by the following equation suggested by Shigesato and Paine[32] for the ITO with different tin concentrations.

$$V_\mathrm{F} = (3\pi^2)^{\frac{1}{3}}[(h/2\pi)/m^*]n^{\frac{1}{3}} \tag{1}$$

$$\iota = V_\mathrm{F}\tau(3\pi^2)^{\frac{1}{3}}[(h/2\pi)/e^2]n^{\frac{2}{3}}\rho^{-1} \tag{2}$$

Here, n is electron concentration, m^* is effective mass of electron, τ is relaxation time, V_F is electron velocity, and ι is MFP. As shown in the figure, the calculated MFP of room-temperature-deposited IO was less than 7 nm and the MFPs of room-temperature-deposited ITO were less than 6.5 nm. Because the MFPs of the deposited IO and ITO were less than the typical grain size, the grain boundary scattering could be ignored under these experimental conditions. In the figure, the lower tin concentration appears to show a larger MFP for similar electron concentration, which is due to the tin neutral impurity scattering. It is difficult to determine the relative importance of neutral impurity scattering and ionized impurity scattering, because mobilities and MFPs shown in Figs. 2 and 3 are significantly dependent on both electron concentration and tin wt%. However, the electron concentration appears to be more important than tin wt% at the low tin percentages, even through the effect of tin wt% on mobility increases at the high tin percentages.

The lowest resistivity of IO and ITO deposited by IBAE was $3.6 \times 10^{-4}\ \Omega\cdot\mathrm{cm}$ for IO, 4×10^{-4}–$5 \times 10^{-4}\ \Omega\cdot\mathrm{cm}$ for 5–15 wt% SnO_2, and $7 \times 10^{-4}\ \Omega\cdot\mathrm{cm}$ for 25 wt% SnO_2. The electrical characteristics of IO and ITO deposited at room temperature by IBAE were more dependent on the electron concentration than tin wt% at the low tin percentages. At the high

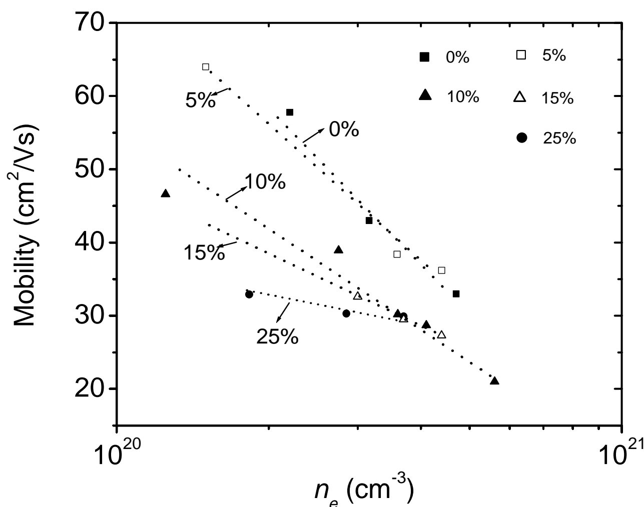

Fig. 2. Hall mobility of amorphous IO and ITO thin films as a function of free electron concentration for various tin oxide weight percentages.

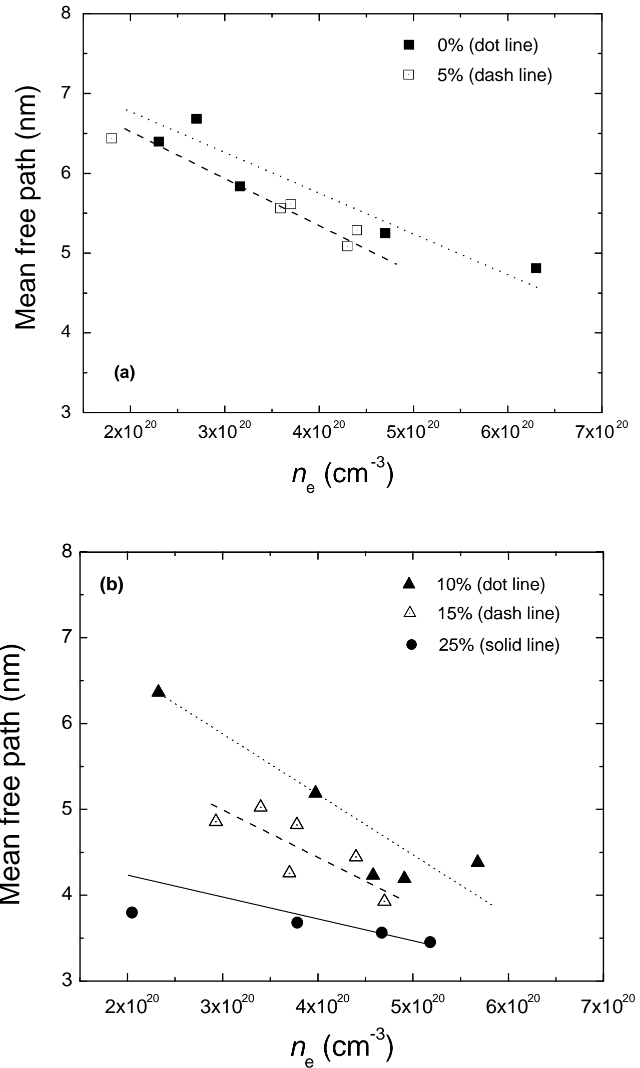

Fig. 3. Mean free path of amorphous IO and ITO thin films as a function of free electron concentration for various tin oxide weight percentages.

tin percentages, neutral scattering by tin atoms also affected mobility and conductivity, therefore, for the tin wt% investigated, important scattering mechanisms appear to be related to the ionized impurity scattering by oxygen vacancy and the neutral impurity scattering by neutral tin atom.

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