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ScienceDirect
Scripta Materialia 60 (2009) 48–51
Scripta MATERIALIA
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Influence of radio frequency sputtering power towards the properties of indium zinc oxide semiconducting films

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Received 23 July 2008; revised 14 August 2008; accepted 28 August 2008
Available online 13 September 2008

We studied the influence of radio frequency (r.f.) power of a sputtering system towards the physical, electrical and optical properties of IZO semiconducting film with an Ar/O_2 gas mixture. Nodules are present when sputtered at 1.8 W cm^{-2}. Resistivity is lowest and mobility highest at low r.f. power. The $Zn_2In_2O_5$ crystallite influences the film's resistivity and mobility. The optical transparency is >80%, while the optical band gap ranges from 3.0 to 3.15 eV and is dependent to the r.f. power.

Keywords: Indium zinc oxide; IZO; R.f. magnetron sputtering

Wide band gap transparent conducting oxides have provoked great interest recently as they have many potential applications, including transparent conducting electrodes [1] and sensors [2]. The development of transparent oxides offers flexibility for the room temperature fabrication of devices having low voltage operations, and the design of such devices will pave the way to the next generation of display technologies based on transparent and flexible devices. A good transparent conducting oxide should have a wide optical band gap, good electrical conductivity (σ) and high optical transparency [3]. Many transparent conducting oxides, composed of combinations of In, Zn, Cd, Sn and Ga, have been widely used in optoelectronic devices. However, indium zinc oxides (IZO) have attracted great attention because of their excellent optical transmission, high σ, chemical stability, thermal stability and low compressive stress. A ternary In_2O_3–ZnO film has been shown to be a promising material due to its higher σ, higher optical transparency, smoother surface [4–6] and higher etch rate [7] in comparison to ITO thin films. The In_2O_3–ZnO system can be subdivided into three groups: (i) In_2O_3-rich, (ii) homologous and (iii) ZnO-rich [8,10]. Kasper [9] demonstrated the formation of the five homologous compounds of $Zn_kIn_2O_{k+3}$ ($k = 2$–5, 7) and Moriga et al. [10] identified nine $Zn_kIn_2O_{k+3}$ homologous compounds with $k = 3, 4, 5, 6, 7, 9, 11, 13$ and 15, but did not include $k = 1$ or 2 as they are unstable in the bulk phase.

Several techniques, such as magnetron sputtering [11–14], pulse laser deposition [15], sol-gel processing [16] and spray pyrolysis [17], have been used to create IZO thin films. In this work, we investigate the effect of radio frequency (r.f.) power on the electrical, optical and physical properties of IZO semiconducting films prepared by r.f. magnetron sputtering.

IZO thin films were prepared on quartz substrate by r.f. magnetron sputtering. The quartz substrate was ultrasonically cleaned first in acetone and then ethanol for 20 min each. Prior to introducing a reaction gas, the chamber was evacuated down to a pressure of 4.4×10^{-6} Torr. The films were prepared using an Ar/O_2 (90/10 vol.%) gas mixture at a pressure of 10 mTorr. A single IZO target with In_2O_3 and ZnO in a molar ratio of 1:1 (atomic ratio Zn:(Zn + In) = 1/3) was used as it has been reported [18] to produce the highest charge mobility (μ). The diameter of the target was 3 in. The deposition of IZO film was performed at three different values of r.f. power: 1.8, 3 and 6 W cm^{-2}. The target–substrate distance during the deposition was kept at 15 cm. Oxygen was used to intentionally increase the electrical resistivity (ρ) of the film as we were not interested in creating a high conducting oxide as electrical electrodes but instead aimed to produce semiconducting films for active devices such as transistors. The temperature of the substrate was maintained at room temperature with a water cooling system.

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doi:10.1016/j.scriptamat.2008.08.039

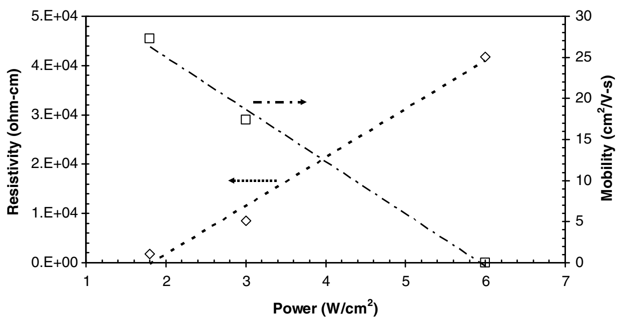

Figure 1. Electrical resistivity and Hall mobility of IZO deposited at different r.f. powers.

Films of various thicknesses, ranging from 25 to 490 nm, were fabricated and characterized.

The film thickness of IZO film and the surface morphology were measured using an atomic force microscope (AFM). X-ray diffraction (XRD) data were obtained by using Cu K_α radiation on a Rigaku *R*-axis Rapid diffractometer. The atomic ratio of the deposited IZO film was determined by energy-dispersive X-ray spectrometry (EDX) attached to field-emission gun scanning electron microscope (SEM). The ρ was measured using a 4-point probe system. The μ of the as deposited film was measured with a Hall system (ECOPIA Hall effect measurement system HMS 3000).

The deposition rates of the IZO film at different sputtering powers of 1.8, 3 and 6 W cm^{-2} were 0.14, 1.13 and 2.72 nm min^{-1}, respectively. This is obviously because more ions in the plasma are formed as the r.f. power increases.

Figure 1 shows the measured ρ and Hall μ for IZO films deposited at different r.f. power. The μ measurement for film deposited with r.f. power of 6 W cm^{-2} did not yield any reading and so was assumed to be 0 cm^2 V^{-1} s^{-1}; this could be due to the film's high ρ. Therefore, increasing the r.f. power to a very high value to reduce deposition time will compromise the film's σ. It was reported in Refs. [19,20] that thicker films exhibit lower ρ, which is in contrast with our result, where thicker films had a higher ρ (e.g. for 3 W cm^{-2}, $\rho = 1.2 \times 10^4\ \Omega$ cm at 70 nm and $2.1 \times 10^4\ \Omega$ cm at 200 nm). Therefore, the previously reported ρ drop with increasing film thickness does not apply for this sputtering condition and contradicts the theory of quantum confinement. As reported by Cho et al. [21], the ρ increases with sputtering power, suggesting more structural defects in the films as a result of higher r.f. power, which might not be valid in our case as the sputtering used by Cho et al. was much higher (8.2 W cm^{-2}).

The atomic ratio (*M*) of individual constituents (Zn:(Zn + In)) was determined by EDX. The measurements revealed that atomic ratios of the films deposited at the different applied powers are relatively constant at approximately 3/4. It was reported in Ref. [22] that the μ of the film decreases as *M* increases. In that study two targets were used (ZnO and In_2O_3) with varied applied powers (co-sputtering) to achieve different *M*. However, in our case only one target with a molar ration of 1:1 In_2O_3:ZnO was used, and hence, as expected, the atomic ratio of individual constituents was found to be similar for all films sputtered at the different r.f. powers. The film's *M* of 3/4 suggests that this sputtering favors the growth of Zn in preference to In, even though a sputtering target with an *M* of 1/3 was used.

The surface morphology of the IZO film deposited at different r.f. power is shown in Figure 2. The presence of circular nodules on the IZO film sputtered with an r.f. power of 1.8 W cm^{-2} is significant, while at 3 W cm^{-2} only some irregular shapes nodules are present. At 6 W cm^{-2}, however, these nodules are absent. The nodules are approximately 9–10 nm in height, with a diameter of approximately 1 µm in Figure 2(a) and 0.5 µm in Figure 2(b). Low r.f. power sputters at a lower rate and could give rise to nodules due to sub-oxide on the target surface [23]. The r.m.s. roughness of the films over a 4 µm × 4 µm area deposited at 1.8, 3 and 6 W cm^{-2} was 2.26, 2.16 and 0.1 nm, respectively. Although film deposited at high power has a high ρ, it produces a significantly smoother film.

XRD was used to observe the structural changes of the IZO films deposited at different applied powers. From Figure 3, it is obvious that the structure of the deposited film is polycrystalline. From the XRD spectra one can see two peaks for IZO homologous compound, i.e. peak #1 for $Zn_2In_2O_5$ (008) and peak #2 for $Zn_4In_2O_7$ (0012); peak #3 is for the In_2O_3 (400) structure. The ratios between the different peaks of Figure 3 are tabulated in Table 1. From the results, it can be deduced that the intensity ratio of peak #1 with respect to peaks #2 and #3 changes with different r.f. power, while the ratio between peak #2 and peak #3 remains relatively constant. As was reported in Ref. [22], the μ and σ of the $Zn_kIn_2O_{k+3}$ homologous compound are strongly dependent on the *M* of the deposited film,

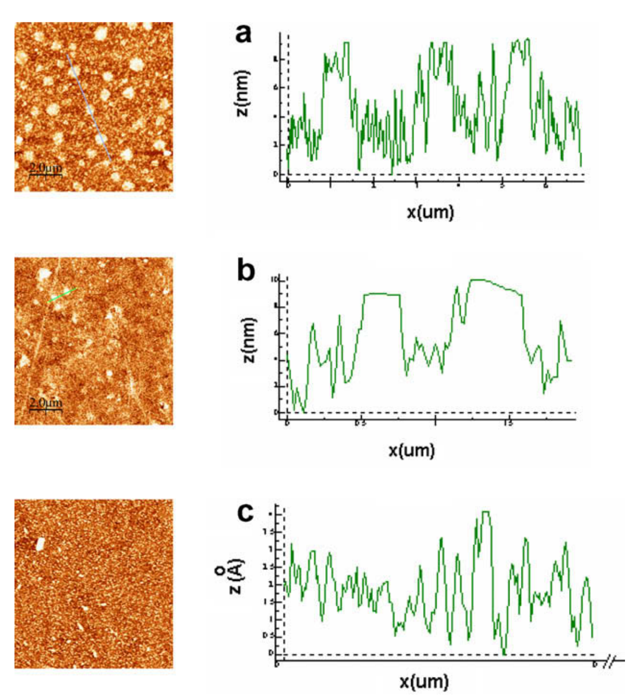

Figure 2. AFM images of IZO films deposited at (a) 1.8 W cm^{-2}, (b) 3 W cm^{-2} and (c) 6 W cm^{-2}.