Current Applied Physics 10 (2010) S506–S509

Contents lists available at ScienceDirect

Current Applied Physics

journal homepage: www.elsevier.com/locate/cap

rf-Magnetron sputtered ITO thin films for improved heterojunction solar cell applications

Vinh Ai Dao [a], Hyungwook Choi [a], Jongkyu Heo [a], Hyeongsik Park [a], Kichan Yoon [a], Youngseok Lee [a], Yongkuk Kim [a], Nariangadu Lakshminarayan [c], Junsin Yi [a,b,*]

[a] School of Information and Communication Engineering, Sungkyunkwan University, Republic of Korea
[b] Department of Energy Science, Sungkyunkwan University, Republic of Korea
[c] Department of Physics, Madras Christian College, Chennai 600 059, India

ARTICLE INFO

Article history:
Received 1 November 2009
Received in revised form 11 January 2010
Accepted 16 February 2010
Available online 19 February 2010

Keywords:
Indium tin oxide
rf Sputtering
Substrate temperature
Heterojunction solar cells

ABSTRACT

Indium tin oxide (ITO) films of low resistivity, high transmittance and good figure of merit were prepared by radio frequency magnetron sputtering, at different substrate temperatures (T_s) under such a high λ/d value and used as anti-reflection layer in heterojunction solar cells. For film deposition in the T_s range 150 °C < $T_s \leqslant$ 250 °C, XRD shows that coexistence of the $\langle 1\,0\,0\rangle$ and $\langle 1\,1\,1\rangle$ textures. The resistivity and Hall mobility of ITO films were improved due to thermally induced crystallization. However, carrier concentration of these ITO films is sensitive to the T_s. We attributed these effects to the Ar^+ ions bombardment and differing adatom mobility of the heated atoms on the substrate under such a high λ/d value. Those ITO films were used to fabricate single-side heterojunction solar cells. As the T_s is increased, the device performance improves and the best photo voltage parameters of the device were found to be V_{oc} = 640 mV, J_{sc} = 36.90 mA/cm^2, FF = 0.71, η = 16.3% for T_s = 200 °C. The decrease in performance beyond the T_s of 200 °C is attributed to hydrogen effusion to the defect in emitter layer. We noted that the figure of merit value of ITO films was reflected in the performance of devices.

1. Introduction

Due to their unique functional properties, transparent conducting oxide (TCO) thin films have been extensively studied and utilized in electronic and optoelectronic device fabrication technologies such as organic light emitting diodes (OLEDs) [1], liquid crystal displays (LCDs) [2], plasma panel displays (PDPs) [3] and solar cells [4]. One of such useful materials is ITO (indium tin oxide). The current application of ITO films emphasizes the requirement of achieving the lowest possible electrical resistivity with the optimized highest transparency in the visible range [5]. The electrical and optical properties of ITO thin films are critically process dependent. The transparency and conductivity of highly degenerate and wide band gap oxide semiconductor films can be varied by adjusting the deposition conditions. The ability of depositing highly conductive and transparent ITO films is a key issue to obtain the best performance of solar cells. A large conductivity in an ITO film is accompanied by larger light absorption and opacity [6]. Therefore, an optimized performance is sought for an effective application to solar cell fabrication.

Earlier, most research groups have studied the effects of the deposition condition, such as substrate temperature (T_s), rf power, oxygen-to-argon ratio, deposition pressure, substrate-to-target distance and bias voltage, on the properties of ITO [7,8]. However, the influence of the substrate temperature during deposition under high λ/d value on the properties of ITO films, and hence on the performance of heterojunction solar cells, has not been reported.

In this paper, we report the effects of the T_s on the properties of ITO films, which are deposited by rf-magnetron sputtering, and hence on the performance of the related heterojunction solar cells. At first, the properties of ITO films such as structure, electrical properties, figure of merit and optical properties were characterized and discussed with respect to the substrate conditions. We then discuss the performance of heterojunction solar cells as related to the effects of the T_s on the ITO films used in these cells.

2. Experimental details

ITO films were deposited from ITO target containing 90 wt.% of In_2O_3 and 10 wt.% of SnO_2 using the rf-magnetron sputtering technique. Commercially available polished microscopic glass slides were used as the substrates. The substrates were thoroughly cleaned in acetone, isopropyl alcohol and de-ionized water baths, in the given order. Using an oil diffusion pump, the chamber of

* Corresponding author. Address: School of Information and Communication Engineering, Sungkyunkwan University, Republic of Korea. Tel.: +82 31 290 7139; fax: +82 31 290 7179.
E-mail address: yi@yurim.skku.ac.kr (J. Yi).

1567-1739/$ - see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.cap.2010.02.019

the sputtering unit was evacuated to a pressure of 10^{-5} Torr before admitting argon (99.999% purity) at a pressure of 2×10^{-3} Torr. Before deposition, the target was treated with pre-sputtering for 10 min in order to normalize the initial sputtering condition of the target surface, during which the substrates were prevent from being exposed to the target; with the aid of a shutter. The rf power density was 0.9 W cm^{-2}. The T_s was varied from room temperature to 250 °C and was measured by a thermocouple attached to the backside of substrate. The space between target and substrate was fixed at a short distance of 2.6 cm to ensure that $\lambda/d > 0.8$, where λ the free path length of sputtered particles is appropriate to the working pressure and d is target–substrate spacing. The sputtered particles can reach the films without being scattered or with the least scattering and the energy is relatively high. Films prepared under such a condition are expected to have different properties from those prepared at low λ/d value [8].

The thickness measurements of ITO thin films were performed by using ellipsometry (SE MF-1000), and the average value was found to be 100 ± 5 nm. The structure properties of ITO thin films were analyzed with X-ray diffraction (XRD) profiles by Bruker AXS D8 Discover using Cu K_α radiation (λ = 0.15,418 nm). The sheet resistance was measured with a four-point probe. The resistivity (ρ), the carrier concentration (n) and Hall mobility (μ) of the films were measured at room temperature with using the Hall Effect measurement done using the van der Pauw geometry. The optical transmittance was measured in the wavelength range of 300–2000 nm using UV–Vis–NIR spectrophotometer.

The heterojunction devices were fabricated by the deposition of the intrinsic hydrogenated amorphous silicon (a-Si(i):H) layer and *p*-type hydrogenated amorphous silicon (a-Si(p):H) as an emitter on the polished side of the *n*-type Czochralski (CZ) Si substrates. Details about the preparation can be found in Ref. [9], The thickness of the a-Si(i):H layer was around 5 nm. The thickness of the a-Si(p):H layers was fixed at 7 nm. The ITO deposition was performed using a metal mask that was directly placed on the a-Si:H surface to form square-shaped ITO layers on the a-Si:H. Ag/Al and Al electrodes (∼1000 Å) were formed on the ITO and back surfaces by evaporation to make a good ohmic contact. Reactive ion etching (RIE) was carried out using SF_6 gas on the top side for mesa-etching. Finally, before the measurement of the solar cell characteristics, the above sample was annealed in air environment at 100 °C (1/2 h) to improve the solar cell performance. The solar cells were characterized by light current density–voltage under AM 1.5, 100 mW cm^{-2} condition at 25 °C.

3. Results and discussion

The X-ray diffraction patterns of ITO films deposited at different T_s are shown in Fig. 1. The occurrence of crystalline phase starts at room temperature and the films show the presence of a weak crystalline structure with ⟨1 0 0⟩ orientation, in a disordered background. Increasing T_s results in a slightly improved crystallization and the emergence of a (2 2 2) diffraction peak. As T_s is raised further, the intensity of the (4 0 0) peak increases relative to that of (2 2 2) peak indicating the coexistence of the ⟨1 0 0⟩ and ⟨1 1 1⟩ textures. Considerable parts of these phenomena have also been reported by other studies. The (2 2 2) peak becomes dominant with an increased oxygen flow rate [5] and an increased substrate temperature [10,5] which seems to agree with our data. Further, the crystallization of the film along (4 0 0) preferred plane is sensitive to the deposition conditions and becomes thermodynamically favorable [5] with higher mobility of adatoms on the substrate. As well known that the diffusion length, Λ, of the adatom in time t can be expressed as follows [11]:

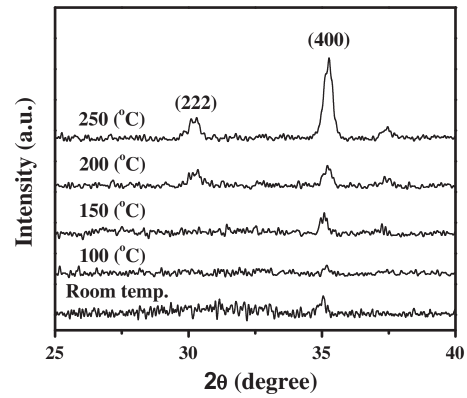

Fig. 1. X-ray diffraction patterns of ITO films deposited at different substrate temperatures.

$$\Lambda = 2(Dt)^{1/2} = v_{os}^{1/2} a (e^{-E_s/KT})^{1/2}, \tag{1}$$

where D is the diffusivity or diffusion coefficient of the adatoms on the substrate of the films, v_{os} is the vibrational frequency of adatoms, a is the atomic dimension and E_s is the reaction activation energy of surface diffusion. From Eq. (1) it is obvious that if the adatoms that are on substrate had a sufficient amount of energy, then the ITO films would be oriented along the thermodynamically favorable ⟨1 0 0⟩ direction. As well known, the average energy of adatoms is considered to be determined by the kinetic energy of the sputtered atoms just before arriving at the substrate and the substrate heating that imparts a thermal energy to the heated atoms which enhances the adatom mobility. Consequently, the ITO films would have a higher portion of (4 0 0)-oriented nuclei as the substrate temperature is increased.

The density of interstitial oxygen $[O_i]$ that depends on the T_s was calculated by Mergel and Qiao [12] as:

$$[O_i] = 4.5 \times 10^{20} cm^{-3} \frac{\varepsilon_{net}}{1\%}. \tag{2}$$

where ε_{net} is a net lattice expansion and $\varepsilon_{net} = \Delta d/d_o$ with $\Delta d = d - d_o$. The $[O_i]$ is displayed as a function of the T_s as shown in Fig. 2. It can be seen that the $[O_i]$ decreases with increasing substrate temperatures up to 200 °C, and it then slightly increases with the further increase of T_s. In this investigation we attribute the change of the $[O_i]$ to the Ar^+ ions bombardment and the different adatom mobility on the substrate under different T_s and such a high λ/d condition. With the increase of T_s, the adatoms mobility of the

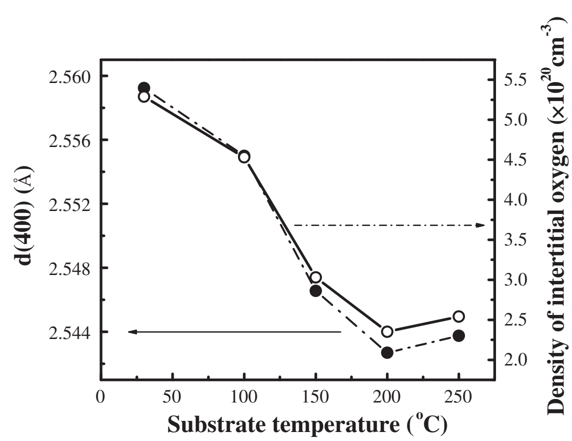

Fig. 2. The variation of lattice distances (d) of (4 0 0) planes and density of interstitial oxygen with substrate temperature.