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Electrical properties of vacuum-annealed titanium-doped indium oxide films

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ABSTRACT

Titanium-doped indium oxide (ITiO) films were deposited on Corning glass 2000 substrates at room temperature by radio frequency magnetron sputtering followed by vacuum post-annealing. With increasing deposition power, the as-deposited films showed an increasingly crystalline nature. As-deposited amorphous ITiO films obtained at 20 W began to crystallize at the annealing temperature of 155 °C. Although there was no significant change in the crystalline structure of the films, electron mobility improved gradually with further increase in the annealing temperature. After post-annealing at 580 °C, the highest electron mobility of 50 $cm^2 V^{-1} s^{-1}$ was obtained. Compared with the amorphous ITiO films, the ITiO films with a certain degree of crystallinity obtained at high deposition power were less affected by the vacuum annealing. Their electron mobility also improved due to post-annealing, but the increase was insignificant. After post-annealing, the optical transmission of the 325 nm-thick ITiO films showed approximately 80% at wavelengths ranging from 530 to 1100 nm, while the sheet resistance decreased to 10 Ω/sq. This makes them suitable for use as transparent conductive oxide layers of low bandgap solar cells.

1. Introduction

Transparent conducting oxide (TCO) films are widely employed in optoelectronic devices, such as flat panel displays, solar cells, smart windows, light-emitting diodes, and touch panels [1]. Good quality TCO films have excellent transmittance (>85%) in visible wavelengths, as well as electrical conductivity ($<10^{-4}$ Ω cm). In recent years, TCO films are required to have high electron mobility and low carrier concentration, which can increase optical transmission in the long wavelength region without sacrificing electrical conductivity.

Among the high mobility TCO films, thin films doped with Mo, W and Ti show excellent performance. The reported mobility changed from tens to hundreds according to different deposition techniques and experimental parameters [2–4]. Samples from the pulsed laser deposition showed higher mobility than those obtained by other deposition techniques including spray pyrolysis, direct current sputtering, radio frequency (RF) sputtering, evaporation, channel spark ablation, and hollow cathode sputtering [5–7]. In most deposition processes, obtaining perfect crystallization and enhancing electron mobility involve maintaining the temperature of substrates at values larger than 300 °C (at times >500 °C).

In the present work, ITiO films are deposited on Corning glass 2000 substrates at room temperature by RF magnetron sputtering using a 1 wt.% TiO_2-doped In_2O_3 target. The effects of the processing parameters on the characteristics of the ITiO films are studied, and a subsequent post-annealing in vacuum with fast heating rate to improve film performance is also investigated.

2. Experimental

ITiO thin films were prepared on Corning 2000 glass substrates by RF magnetron sputtering in the deposition system, "Zeester," at room temperature from 1 wt.% TiO_2-doped In_2O_3 target (AJA International, Inc.). The working pressure during the deposition was controlled at 1.8 μbar by adjusting a pure argon flow. The deposition power was varied from 20 to 150 W. The as-deposited ITiO thin films were post-annealed in a vacuum oven (at 2–3 × 10^{-3} Pa) for 1 h at different temperatures (heating rate = 60 °C/min).

Carrier concentration, hall mobility and resistivity were obtained from the Hall Effect measurements (PhysTech GmbH RH2030). The transmittance was measured by Fourier transform photocurrent spectroscopy (FTPS) at wavelengths ranging from 530 to 1780 nm. The crystalline structure of the films were characterized by X-ray diffraction (XRD) (PHILIPS PW 1729 X-ray generator), and the diffractometer was operated at 40 kV and 20 mA.

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3. Results and discussion

3.1. The effect of power on the structure and electrical conductivity of ITiO films

Generally, ITiO films obtained in high pressure argon plasma have dark appearance and poor optical properties. By introducing oxygen during film growth, the transparency of the films increases but conductivity is greatly reduced. Thus, all the ITiO films in our work were deposited at low pressure pure argon, which was about 0.18 Pa. The deposition power was changed from 20 to 150 W.

Fig. 1 shows the XRD patterns of the as-deposited ITiO films obtained at different power levels. The data were smoothed with a 5 pt filter. The film obtained at the low deposition power was amorphous, and the films gradually became crystalline with increasing power. The ITiO films were amorphous at 20 W. The films gradually crystallized as the power increased to 50 W. The crystalline peaks became very pronounced at 75 W. As the RF power increased, the number and momentum of both the argon ions and the sputtering particles also increased. The atoms became more mobile on the surface of the film as the result of higher bombardment rates on them. This added mobility to the film atoms helps release stress from the films, facilitating further crystallization [8]. All the peak positions were found to be in good agreement with crystalline indium oxide, and no extra peaks due to the addition of titanium in the indium oxide films were observed.

Fig. 2 gives the electron mobility μ (squares), carrier concentration n (triangles), and resistivity ρ (circles) of the as-deposited ITiO films from Hall Effect measurements. As-deposited ITiO films obtained at a low deposition power showed low mobility and high carrier concentration. With increasing deposition power, carrier concentration gradually decreased, and fluctuations in mobility were observed. The reduction in carrier concentration was greater than the increase in mobility. Therefore, with increasing deposition power, resistivity increased gradually.

ITiO film deposited at 20 W had the lowest resistivity, and its mobility was higher than those deposited at 30, 40, and 50 W. Thus, we selected this film for the present study.

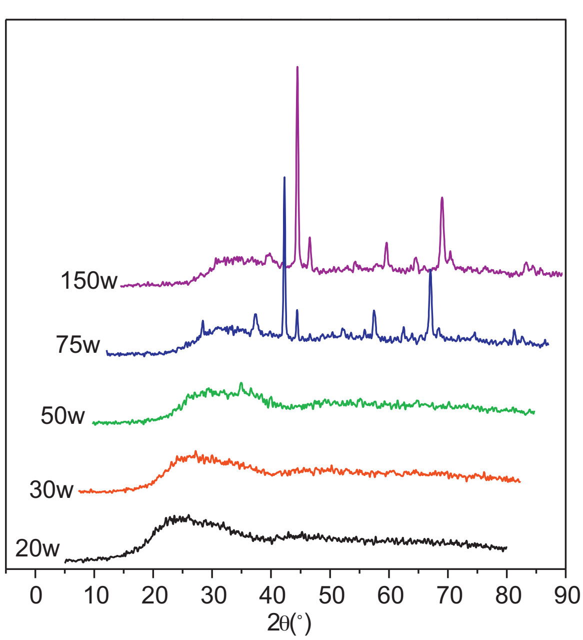

Fig. 1. XRD patterns of as-deposited ITiO films at different powers.

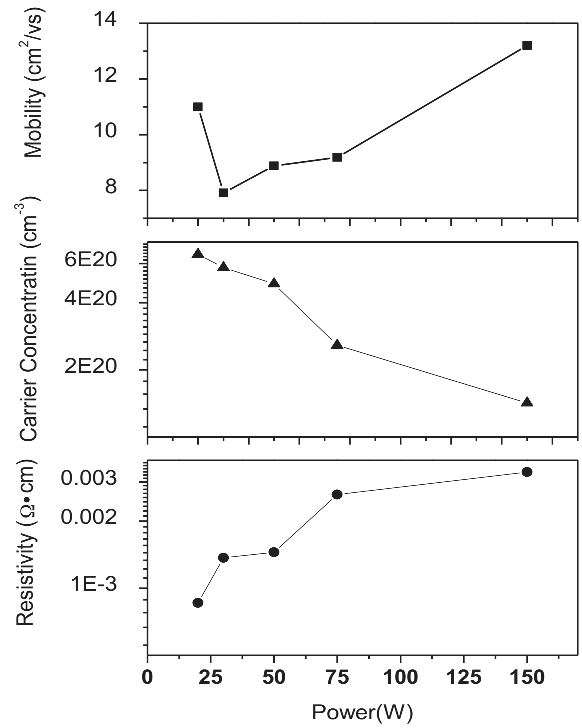

Fig. 2. Electrical properties of as-deposited ITiO films at different deposition powers.

3.2. The effect of annealing temperature on the structure and electrical properties of ITiO films

As-deposited ITiO films obtained at 20 W were post-annealed in a vacuum ($2–3 \times 10^{-3}$ Pa) for 1 h at various temperatures (heating rate = 60 °C/min). Fig. 3 shows the XRD patterns of ITiO films after post-annealing in a vacuum at different temperatures. The data were smoothed with a 5 pt filter. Although amorphous ITiO films started to crystallize at 155 °C, the peak intensity did not increase further with increasing annealing temperature. All diffraction lines were assigned to In_2O_3, although the relative intensity of each peak was different with different post-annealing temperatures. Given that the film was too thin (about 80 nm), it appeared as a broad peak in the background of the XRD patterns.

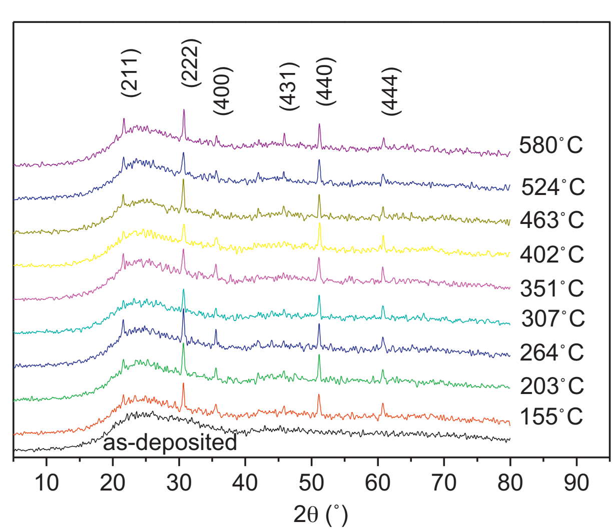

Fig. 3. XRD patterns of as-deposited ITiO films and after post-annealing in vacuum at different temperatures.