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ScienceDirect
Applied Surface Science 254 (2008) 2250–2254
applied surface science
www.elsevier.com/locate/apsusc

Growth of transparent conducting nano-structured In doped ZnO thin films by pulsed DC magnetron sputtering

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Received 10 May 2007; received in revised form 3 July 2007; accepted 5 September 2007
Available online 8 September 2007

Abstract

Transparent conducting nano-structured In doped zinc oxide (IZO) thin films are deposited on corning 7059 glass substrates by bipolar pulsed DC magnetron sputtering with variation of pulsed frequency and substrate temperature. Highly *c*-axis oriented IZO thin films were grown in perpendicular to the substrate on the 30 kHz and 500 °C. The IZO films exhibited surface roughness of 3.6 nm similar to the commercial ITO and n-type semiconducting properties with electrical resistivity (carrier mobility) of about 5×10^{-3} Ω cm (14 cm^2/V s). The optical characterization showed high transmittance of over 85% in the UV–vis region and exhibited the absorption edge of near 350 nm. In micro-Raman spectra, the origin of two additional modes is attributed to the host lattice defect due to the addition of In dopant. These results suggest that the IZO film can possibly be applied to make transparent conducting electrodes for flat panel displays.

Keywords: Nano-structured; Pulse DC magnetron sputtering; In doped ZnO (IZO); Transparent conducting oxide (TCO)

1. Introduction

Transparent and conducting oxide (TCO) films with unique characteristics of low resistivity and high transparency over the visible wavelength region have numerous applications in optoelectronic devices including thin film solar cell [1], organic light emitting devices (OLED) [2], and other flat panel displays (FPDs) [3]. Especially, tin doped indium oxides (ITO) are used as anodes of several devices because of their high conductivity and transparency over the visible range and their high work function. Tin doped indium oxides (ITO) have been mainly used as anodes in FPDs because of their high conductivity and transparency over the visible range and their high work function. However, the price of indium is increasing drastically due to the high demand of ITO in the rapid development of FPDs industry. Also, the toxic nature and high cost due to the scarcity of indium have led researchers to seek an alternative candidate for ITO [4]. Recently, In or Al doped zinc oxide (IZO or AZO) films [5,6] have been considered as possible alternatives to ITO films because ZnO thin films are more stable against hydrogen plasma, more abundant, and less expensive in comparison with the ITO films, which make them appropriate for potential use as anodes in FPDs. ZnO-based films were deposited by several techniques such as radio frequency (RF) sputtering process [7], pulsed laser deposition (PLD) [8], sol–gel method [9], and chemical vapor synthesis (CVS) [10]. Recently, bipolar pulsed DC magnetron sputtering process [11] has attracted notice as the deposition method of the industrially important coatings because it has higher deposition rate of defect-free ceramic films than RF magnetron sputtering process. Particularly, it can also relax the occurrence of arc events at the oxide targets involved in the continuous DC sputtering process. There was a previous report on bipolar pulsed DC magnetron sputtering deposition of AZO thin films [12]. However, reports on bipolar pulsed DC magnetron sputtering deposition of IZO thin films are hard to find.

In the present work, IZO thin films were prepared by bipolar pulsed DC magnetron sputtering on corning 7059 glass

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doi:10.1016/j.apsusc.2007.09.008

substrates. Our experiment reports on the physical properties of IZO thin films with film thicknesses used as anodes in FPDs. The structural, the optical, and the electronic properties of the IZO films have been investigated with various pulsed DC parameters and substrate temperature.

2. Experimental details

IZO films were deposited on corning 7059 glass by asymmetrical bipolar pulsed DC magnetron sputtering. The targets were prepared by sintering the mixing 99.99% ZnO and In_2O_3 powders with In fraction of 1 at%. The sputtering chamber was pumped down to 1×10^{-6} Torr by turbo molecular pump. The substrate to target distance, sputtering time, working pressure, and pulsed DC power were kept at 7 cm, 15 min, 5 mTorr, and 150 W, respectively. The pulsed frequency of power and the substrate temperature maintained during the sputtering process were varied in the 10–70 kHz at substrate temperature (T_S) of 500 °C and in 100–500 °C at DC pulsed frequency of 30 kHz, respectively. The thicknesses of the deposited films determined by the field emission scanning electron microscopy (FESEM) were in the 200 ± 10 nm range.

The crystal structure, the electrical properties, and the surface roughness of the deposited IZO films were investigated by X-ray diffraction (XRD) measurements, Hall-effect measurements using van der Pauw method, and atomic force microscopy (AFM) at the room temperature. The optical transmittance was recorded using an ultraviolet-visible spectrometer in the wavelength range of 200–1100 nm at the room temperature and calculated the optical band gap energy. The micro-Raman measurements were performed in the back-scattering geometry. The radiation of 514 nm from an Ar laser was focused to ~10 μm in diameter on the samples at room temperature.

3. Results and discussion

Fig. 1 shows the *c*-axis lattice constant and the full-with at half-maximum (FWHM) from the (0 0 0 2) peak of IZO films, calculated by XRD results as a function of pulse frequency. As the pulse frequency increases, the *c*-axis lattice constant slightly increased. The value of FWHM appeared lowest value near the pulse frequency of 30 kHz and it increased at a pulse frequency higher than 50 kHz. Since the value of FWHM is inversely proportional to the crystallite size, it implies that crystallite size increases with the increase in film pulse frequency up to 40 kHz. The increase of the crystallite size and the improvement of the crystallinity are responsible for the decreasing the resistivity due to diminishing in grain boundary scattering.

In Fig. 2(a), the crystalline structure and orientation of the deposited films were investigated by XRD. ω–2θ scans of IZO thin films at various temperatures are plotted. The films were all transparent and well oriented to the *c*-axis without showing any In_2O_3-related peaks in their XRD spectra. All of the films exhibit the preferred *c*-axis orientation due to the minimal surface energy in the ZnO wurtzite structure. To assess the crystallinity of the thin films, FWHM of (0 0 0 2) orientation were measured rocking curves by ω-scan. The inlet is a ω-scan

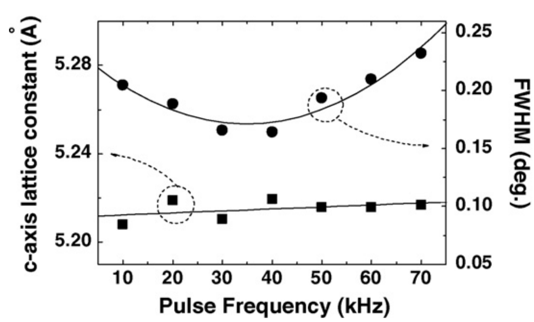

Fig. 1. Variation of the *c*-axis lattice constant and FWHM of IZO films with pulse frequencies.

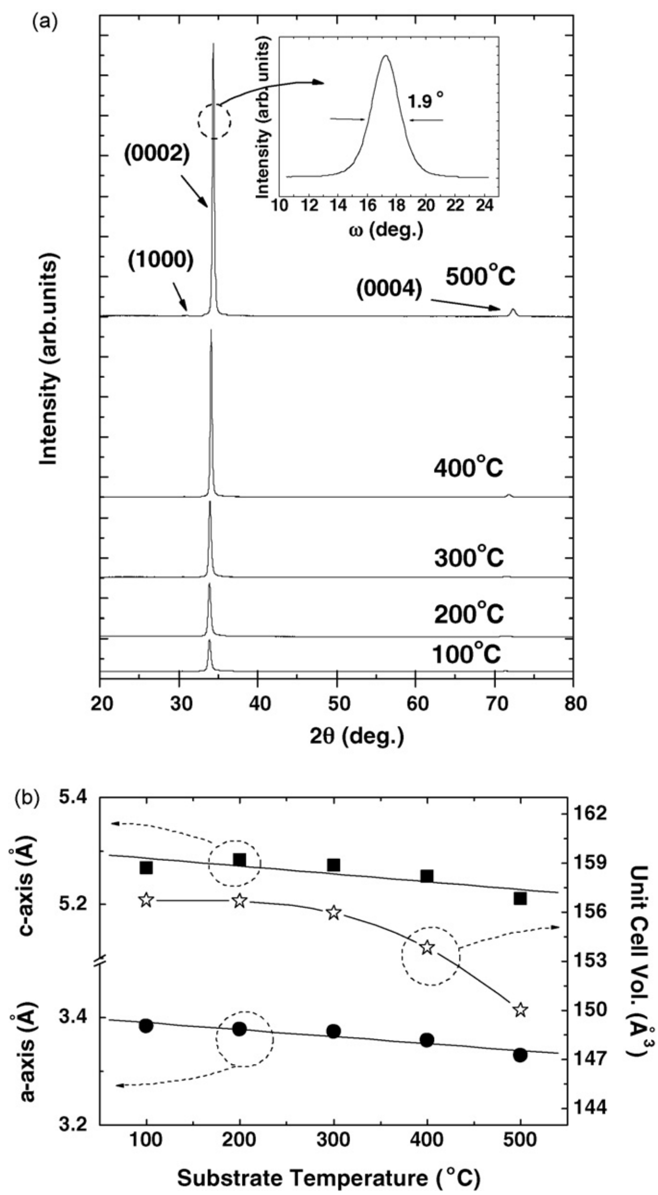

Fig. 2. (a) XRD spectra of IZO films grown on corning 7059 glass substrates with substrate temperatures and the inlet is a ω-scan of IZO film. (b) Variation of the lattice constants and the resultant unit-cell volume of IZO films with substrate temperatures.