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Properties of Zinc Oxide Films Cosputtered with Various Aluminum Contents at Room Temperature

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Abstract--Electrical, optical and material properties of zinc oxide (ZnO) films doped at various aluminum impurities by cosputtered Al and ZnO targets at room temperature has been investigated. The carrier concentration was was found to be apparently increased with the cosputtered Al contents in the ZnO films. At a specific theoretical atomic ratio [Al / (Al +Zn) at.%], a lowest resistivity was achieved for the contribution of the considerable Al donors in the films as well as the superior crystalline structure. In terms of the associated transmittance spectra, the optical window could be effectively enlarged to 3.70 eV due to the Burstein-Moss shift obtained from the cosputtered AZO films at room temperature.

1. Introduction

Transparent and conductive oxide (TCO) films have been widely used for optoelectronic devices. Indium-tin oxide (ITO) is the most commonly transparent electrodes in flat panel displays and solar cells [1-2]. Recently, zinc oxide (ZnO) films have been demonstrated as a promising candidate for altering the commercial ITO films due to their superior chemical selectivity, mechanical properties, abundance in nature, and absence in toxicity. However, undoped-ZnO films are still poor in conductivity. Donor impurities such as III-group elements of indium, aluminum, and gallium incorporated into ZnO films have been demonstrated [3-4]. In addition to the exploration of doped-ZnO materials, the development of the deposition technology at low temperature is also demanded for the upcoming flexible electronic device applications [5-6].

In this paper, for the purpose to prepare AZO films with excellent electrical and optical properties at room temperature, AZO films at various Al doping levels are deposited using a radio frequency (rf) magnetron cosputtering system. ZnO and Al materials are selected as the cosputtered targets to efficiently activate the Al contents introduced in the ZnO films at room temperature. In addition, in contrast with these cosputtered films, AZO films deposited using a specific mixture target (ZnO : Al_2O_3 = 98 : 2 wt.%) are also prepared. The electrical property evolutions included carrier concentration and hall mobility of these cosputtered films are discussed and connected with the corresponding crystalline structure and optical absorption behavior obtained from X-ray diffraction (XRD) patterns and optical transmittance spectra.

2. Experimental procedure

A dual rf power supply (13.56 MHz) with synchronized phase was employed to prepare the undoped ZnO film and AZO films at various Al doping levels at room temperature using ZnO and Al targets. The working pressure was kept at 0.133 Pa under pure argon ambient. The deposition rates of the undoped ZnO and Al films as a function of the supplied rf power are shown in Fig. 1. To prepare ZnO films doped with various Al contents, the rf power supplied to ZnO target was fixed at 320 W while that of Al target were varied from 30 to 70W. According to the molecular weights and the density of ZnO and Al materials, the theoretical atomic ratios (referred to as atomic ratio hereafter) for the cosputtered AZO films were ranged from 7.5 to 15%. Electrical properties were measured by the van der Pauw method. The crystalline structure evolutions were examined by XRD measurements. Optical transmittance spectra were obtained using an UV-Vis-NIR spectrophotometer over a wavelength range from 300 to 800 nm.

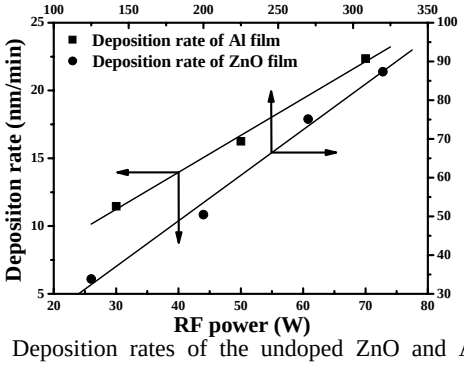

Fig. 1 Deposition rates of the undoped ZnO and Al films as functions of the supplied rf power.

3. Experimental results

Film resistivity, carrier concentration, and hall mobility for the cosputtered AZO films as a function of the rf power supplied on Al target are shown in Fig. 2. The undoped ZnO film deposited at room temperature induced less free electron carriers and resulted in an insulating behavior. With introducing Al impurities in the ZnO films by cosputtering, the carrier concentration was markedly increased with the rf power. A lowest resistivity of 8.79×10^{-3} Ω cm was obtained from the cosputtered AZO film at an atomic ratio of 10%. By contrast, AZO films at a similar Al doping level prepared using a mixture target under an optimal deposition condition at room temperature performed the inferior resistivity of 8.38×10^{-2} Ω cm mainly emerged from the lower carrier concentration. This

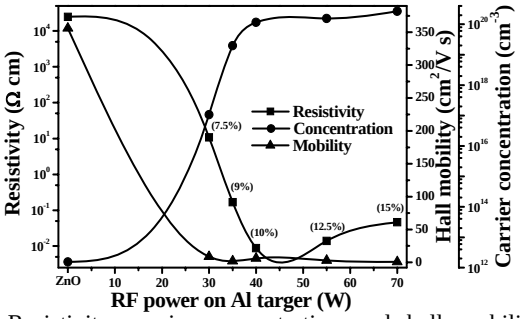

Fig. 2 Resistivity, carrier concentration and hall mobility as functions of the rf power on Al target (the corresponding theoretical atomic ratio indicated in the bracket).

indicated that the doped Al donors could be more effectively activated at room temperature by this cosputtering technology. Although the carrier concentration was continuously increased with the rf power supplied on the Al target, the related film resistivity was gradually increased attributed to the decrease in the hall mobility. The crystalline structures of these AZO films as well as the undoped-ZnO film are shown in Fig. 3. Compared to the undoped ZnO film, the diffraction peak positions of ZnO (002) phase was initially increased with Al contents and then shifted to lower diffraction angles. Since the ion radius of Al was smaller than that of Zn, the diffraction peak shifted toward higher diffraction angle was resulted from the substitution of Zn^{2+} site by Al^{3+} ones. However, at higher Al doping level, those Al ions activated by cosputtering were prone to introduce into the lattice interstitial sites in the ZnO matrix and subsequently enlarged the lattice constant. In addition, it also degraded the grain size growth. As a result, although these cosputtered film possessed higher carrier concentrations due to the substitution of Zn^{2+} site by Al^{3+} as well as the interstitial Al^{3+}, their hall mobility was apparently decreased due to the lattice scattering. As a result, the superior crystalline structure of the cosputtered ZnO film at an atomic ratio of 10% concluded from the narrower FWHM and lower lattice stress in the related diffraction pattern also was responsible for the achievement of the lowest film resistivity. Fig. 4 shows the related optical transmittance spectra (the corresponding optical bandgap derived from the absorption edge also shown in the inset figure). All the cosputtered films performed the excellent optical window around visible wavelengths. In addition, the optical bandgap also had been effectively enlarged to 3.70 eV contributed from the high carrier concentration. The wider optical bandgap of the AZO film prepared by cosputtering technology at room temperature make it more promising for the flexible electronic devices application on the deep-UV wavelengths.

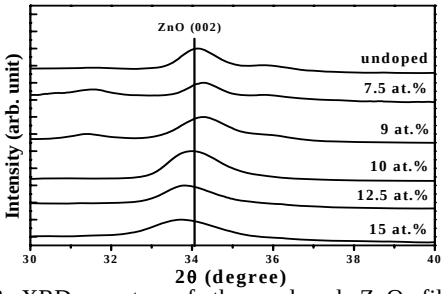

Fig. 3 XRD spectra of the undoped ZnO film and the cosputtered AZO films as functions of atomic ratios.

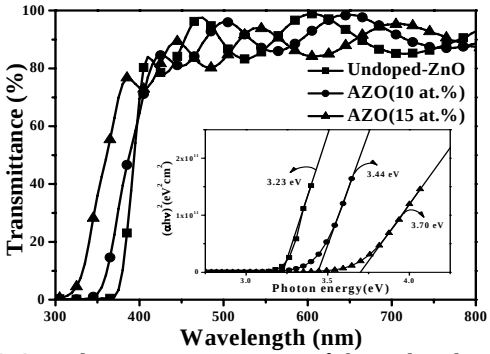

Fig. 3 Optical transmittance spectra of the undoped ZnO film and the cosputtered AZO films at atomic ratios of 10 and 15% (inserted figure is the derivative optical bandgap).

4. Conclusions

Aluminum doped zinc oxide films at various atomic ratios had been successfully prepared by an rf magnetron cosputtering system at room temperature. Compared with the traditional AZO film prepared by a specific mixture target, the cosputtered AZO films performed a much better films resistivity. This great achievement was mainly attributed to the reason that more free carriers were effectively activated by cosputtering deposition even at room temperature. As a result, combined with the consideration on the crystalline structure, a lowest resistivity of 8.79×10^{-3} Ω cm was obtained. Moreover, in terms of the associated transmittance spectra, the optical window also could be effectively enlarged as high as 3.70 eV. The cosputtered AZO films with wider optical bandgap would be much benefit for application on the flexible electronic devices.

Acknowledgments

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