Tailoring of microstructure and optoelectronic properties of Aluminum doped Zinc Oxide changing gun tilt

S. Fernández, J. Grandal, A. Trampert, F.B. Naranjo

ABSTRACT

The influence of gun tilt on microstructure and optoelectronic properties of ZnO:Al thin films deposited by magnetron sputtering onto transparent glass substrates was studied. The structural and morphological film properties were investigated according to the cathode angle that placed in confocal geometry configuration inside sputtering system. X-ray diffraction (XRD) and Atomic Force Microscope (AFM) were used, respectively. Film microstructure was analyzed by high resolution transmission electron microscopy (HR-TEM). Results showed relevant changes in AZO film properties depending on gun tilt, fact that can affect the device performance. Rougher AZO surfaces with lower average transparency values in visible range and worse electrical properties were observed when gun tilt decreased. These strong dependences can be considered as important aspects when design suitable electrodes for optoelectronic devices. In this sense, the ability of tuning AZO film properties as function of geometrical deposition system configuration was evaluated. This approach would allow fabricating coatings based on the same raw material but different optoelectronic properties.

1. Introduction

Aluminum-doped Zinc Oxide (ZnO:Al, i.e. AZO) is being considered as an alternative potential Transparent Conductive Oxide (TCO) material to replace the most common one used so far, Indium Tin Oxide (ITO). These materials are frequently used as transparent electrodes in optoelectronic devices such as flat-panel displays, organic light-emitting devices (OLEDs) and solar cells [1–3]. For such applications, surface morphology of TCO films becomes of paramount importance. Their physical properties must be controlled exhaustively because the device performance can be affected in a critical way. Different types of surface morphologies can be achieved depending on deposition techniques and conditions [4,5].

Among deposition techniques used for AZO films fabrication, magnetron sputtering is considered as the most favourable one. This technique shows many advantages such as the simplicity in fabrication step, low production cost and high level of reproducibility. Finally, the sputtering processes can be performed at low temperature, allowing the use of cheap substrates [6]. Many works report about the strong influence of sputtering deposition parameters such as time deposition (film thickness), substrate bias voltage, substrate temperature, working pressure and radio-frequency (RF) or direct current (DC) power on mechanical and opto-electronic properties of deposited material [8–10]. However, depending on device application, other additional parameters should be also considered. Among them, the geometry of different components inside the sputtering system such as substrate angle and gun tilt can be also relevant. This is because the geometrical configuration, that affects directly the direction of the incoming ion flux of the depositing species, can lead to significant and important changes in the functional properties of the deposited films [11]. In this sense, the manipulation of substrate angle during film deposition, known as oblique-angle deposition (OAD), is emerging as a very useful deposition coating tool used to modify the physico-chemical thin film properties. Using this technique, a wide variety of morphologies can be tailored only changing the direction of the sputtered particles, easily obtained by modifying the substrate angle [12,13]. Films reached under this geometrical configuration can present different surface textures: from columnar to smooth films, and also compactness layers with different degrees of porosity depending on substrate angle [14–16].

Taking into account the paramount importance of deposition system geometry on thin film properties, this work is focused on the study of possible effects on deposited films when the cathode angle is modified. This spatial modification is completely different to what
happens in OAD technique where the substrate angle is modified. The possibility of designing the functional properties of thin films by controlling the physical parameters inside the sputtering system can be considered as a hot topic in the material research field. In this context, to carry out this approach, a sputtering system with confocal configuration is used. The confocal configuration is a good choice to increase the yield of sputtered material and to achieve films with improving film uniformity. In this geometry, the cathode is positioned at an angle relative to normal of substrate surface. At this point, the rotation of substrate is also necessary. The magnetron must be also moved off centre of the rotational axis. The position where the cathode’s centreline intersects with the substrate is changing as cathode angle varies, affecting significantly the properties of thin films [17,18]. In addition, the confocal configuration can reduce the plasma damage because of the remoteness of the sputter gun. In this work, the impact of changing the system geometry by moving the cathode angle on the thin film microstructure, its surface morphology, its compactness, and its opto-electronic properties are evaluated. The relevance of this study is based on the use of this material as front electrode, where its properties can strongly affect quality and stability of subsequent deposited layers of devices. Therefore, the main aim is to establish a correlation between thin film properties and geometrical system configuration to achieve the most appropriate thin film as function of device application.

2. Experimental details

The 0.4 µm-thick AZO thin films were deposited on 15×15 cm² area Corning glass 7059 using a commercial UNIVEX 450B magnetron sputtering system from Oerlikon Leybold Vacuum. This equipment was equipped with four magnetron sources operated by Radio Frequency sputtering system from Oerlikon Leybold Vacuum. The Ar gas used for the deposition process was 99.999%, and the Ar ZnO:Al₂O₃ (98/2 wt%) ceramic target used in this work came from Tosoh Corporation. Prior to AZO deposition, the base pressure was 5 Pa. The deposition parameters were an RF power of 250 W, a substrate temperature of 400 °C and a working pressure of 0.1 Pa, corresponding to 3 sccm of Ar flux. The substrate was rotating at constant rate of 20 rpm. The gun tilt was varied by moving a micrometre ruler from 1.0 to 2.1 cm that corresponded approximately to a variation of angle with respect to normal gun direction of 70–30°, respectively (see pictures in Fig. 1). This was also related to a change in the distance centre of target to centre of substrate 20–8 cm, respectively. At 2.1 cm of gun tilt, the cathode was almost faced up to substrate, point where bombardment of sputtered particles began to be more important.

The structural properties of AZO thin films were characterized by X-Ray diffraction (XRD) using a commercial Panalytical X’Pert MPD X-ray diffractometer with CuKα radiation (λ=1.542 Å). The identification of crystal phase was performed using data from standard table of ZnO (JCPDS 65–3411) [19]. From these measurements, residual intrinsic stress was calculated using biaxial strain model [20] and the following formula, valid for a hexagonal lattice:

\[ \sigma_{film} = \frac{2c_{13}^2 - c_{13}(c_{11} + c_{12})}{2c_{13}} - \frac{c_{film} - c_{bulk}}{c_{bulk}} \]  

where \( c_{ij} \) are the elastic constants of single crystalline ZnO, \( c_{11}=208.8 \), \( c_{13}=213.8 \), \( c_{12}=119.7 \), \( c_{13}=104.2 \) Gpa, and \( c_{film} \) are the lattice parameters of ZnO and the analyzed film, respectively. A negative value of this magnitude denoted a film compressive stress along c-axis. The error bar estimated for this parameter was around 10%.

The surface morphology of AZO films was evaluated using a standard atomic force microscope (AFM) Multimode SPM from Veeco-Digital Instruments operated in tapping mode and an antimony-doped silicon AFM tip from Veeco was used. The roughness was quantified by the average value of the Root Mean Square (RMS) deviation of the AFM measured height from the mean data plane in AFM 10×10 µm² images. The error bar of RMS value due to the roughness variation around the surface was about 15%. AFM images of 1×1 µm² were also taken to analyse the variation of grain size.

The analysis of the film microstructure was performed by transmission electron microscopy (TEM) using a JEOL JEM-3010 microscope. The samples were characterized by conventional, bright field (BF), dark field (DF) and high resolution (HR) TEM. In order to fully characterize the interface, the tilting, twisting and the grain size, specimens were prepared in both cross-section and plan-view configuration using standard methods of mechanical grinding, dimpling, and a final Argon ion milling step until electron transparency was reached.

The sheet resistance \( R_s \) of the samples was measured using a commercial four-point probe system. The film resistivities \( \rho_f \) were obtained from \( \rho_f = R_s t \), being \( t \) the film thickness. Finally, the optical

![Fig. 1. 6/2θ XRD diffraction patterns of AZO thin films deposited at different gun tilt positions. A schematic graphic of geometrical configuration founded inside the sputtering chamber between substrate and gun is included for the extreme positions, 1.0 and 2.1 cm.](image)
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