Applied Surface Science 283 (2013) 623–628

Contents lists available at ScienceDirect

Applied Surface Science

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

The influence of homo-buffer layer on structural, optical and electrical properties of ZnO:Al films

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ARTICLE INFO

Article history:
Received 11 March 2013
Received in revised form 13 June 2013
Accepted 29 June 2013
Available online 20 July 2013

Keywords:
AZO films
Pulsed-laser deposition
Homo-buffer layer
Orthogonal design

ABSTRACT

Aluminum-doped ZnO (AZO) films were fabricated on glass by pulsed-laser deposition based on orthogonal design, and influences of homo-buffer layer on structural, optical and electrical properties of AZO films were investigated. The experiment results demonstrate that the homo-buffer layer is very beneficial to improve the quality of AZO films. In addition, both deposition temperature of homo-buffer layer ($T_{homo\text{-}buffer}$) and deposition oxygen pressure of homo-buffer layer ($P_{homo\text{-}buffer}$) have a considerable influence on the properties of AZO films. By optimizing the deposition parameters of homo-buffer layer and AZO film synthetically, the AZO film with low resistivity ($2.13 \times 10^{-4}\ \Omega\,cm$) and high transmittance (89.1%) has been obtained, which is a promising candidate material in transparent electrode applications.

1. Introduction

Nowadays, indium-tin-oxide (ITO) films are the most-used transparent conductive oxides (TCOs) films for thin-film solar photovoltaic cells, organic lighting diodes, and liquid–crystal displays [1–3]. However, indium is a rare, expensive and toxic metal material. Therefore, it is necessary to develop a new TCO material to replace the ITO [2,4]. Aluminum-doped ZnO (AZO) is a particular promising alternative to ITO for film transparent electrode applications because of its comparable high optical transmittance, low electrical resistivity, inexpensiveness, nontoxicity, and high stability in hydrogen plasma atmosphere [5,6].

To date, various deposition methods including molecular beam epitaxy (MBE) [7], sputtering [8], chemical vapor deposition (CVD) [9], and pulsed laser deposition (PLD) [10] have been widely used to grow ZnO (or doped ZnO) thin films. Among them, PLD is a promising method to prepare high quality metal oxide thin films because the prepared films usually have the same stoichiometry with the target. The deposition process and final performance of metal oxide thin films prepared by PLD are significantly influenced by oxygen pressure, substrate temperature, deposition time, laser energy, etc. [11,12]. Orthogonal design is an effective way to investigate the effects of multiple factors on performance as well as to find the optimal deposition parameters [13,14]. Therefore, we employ the orthogonal design to ascertain the sequence of the influence of factors and optimize the deposition parameters of AZO films.

From the viewpoint of device application, it is desirable to grow AZO films with low resistivity and high transmittance on glass. When AZO film is directly deposited on glass, however, a lot of defects usually exist in the film due to large lattice mismatch and different thermal expansion coefficients [13]. Inserting a suitable buffer layer between the substrate and film is a good way to improve the film performance. Several researchers have improved the quality of AZO films using hetero-buffer layer [15–18]. Compared with the hetero-buffer layer, the homo-buffer layer can avoid the interdiffusion of different atoms and reduce the lattice distortion [13,19]. In this paper, thus we introduce a homo-buffer layer between glass substrate and AZO film. In addition, influences of deposit conditions of homo-buffer layer on the performance of AZO film are investigated.

2. Experiment

The AZO homo-buffer layer and AZO film were deposited on the $1 \times 1\ cm^2$ slide glass substrates by using a pulsed laser deposition system (PLD-5000) with KrF excimer laser (COHERENT COMPEX 205, $\lambda = 248$ nm, 20 ns pulse duration), successively. Prior to deposition, substrates were cleaned in petroleum ether, acetone and methanol for 15 min using an ultrasonic agitator, respectively. The target for both homo-buffer layer and AZO film was ZnO target which was 60 mm in diameter and doped with 2 wt% Al_2O_3 (99.99%

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http://dx.doi.org/10.1016/j.apsusc.2013.06.158

Table 1
The factors and levels of orthogonal experiment for AZO film.

	Factors	Level 1	Level 2	Level 3
A	Oxygen pressure (mTorr)	0	10	50
B	Laser energy (mJ)	300	350	400
C	Substrate temperature (°C)	200	300	400
D	Deposition time (min)	30	45	60

purity). The distance between target and substrate was 87 mm and background pressure of the chamber was 4×10^{-7} mTorr. The composition and crystallinity of deposited films were measured by a Bruker D8 Advance X-ray diffractometer (XRD) using Cu Kα (λ = 1.5406 Å) radiation. The thickness of the films was measured by Filmetrics F50 thin-film analyzer. The Q-Scope 850 atomic force microscope (AFM) was used to characterize the surface roughness of the films. The optical properties of samples were measured by UV–visible spectrophotometer (UV-2550, SHIMADZU). And the electrical properties were measured at room temperature using Hall-effect measurements (HMS-3000, ECOPIA).

In our experiment, we employed the orthogonal design to optimize the basic preparation condition of AZO film, firstly. The oxygen pressure, laser energy, deposition time and substrate temperature were served as the four factors of the orthogonal design as shown in Table 1. The homo-buffer layer was deposited at 100 °C for 5 min in vacuum with laser energy of 400 mJ and repetition rate of 30 Hz. The transmittance was chosen as the evaluation index. And range analysis [20] was used to indicate the effect of each factor and determine the optimal level combination, and the results are shown in Table 2. The factor collocation ($A_1B_3C_2D_2$) corresponding to the largest $\bar{k}_{ij}$ was the optimal level, in which $\bar{k}_{ij}$ was the mean value of the sum of the evaluation indexes of all levels in each factor. The values of R_j of substrate temperature, oxygen pressure, deposition time and laser energy were 3.377, 2.377, 1.480 and 0.478, respectively, in which R_j was the range between the maximum and minimum value of the mean values ($R_j = \max\left\{\bar{k}_{1j}, \bar{k}_{2j}, \bar{k}_{3j}\right\} - \min\left\{\bar{k}_{1j}, \bar{k}_{2j}, \bar{k}_{3j}\right\}$) and means the impact on the transmittance of various factor. Compared with the range values of different factors (R_j), the order of significant factors was: substrate temperature > oxygen pressure > deposition time > laser energy.

In order to investigate the effects of homo-buffer layer deposition conditions on the properties of AZO films, the homo-buffer layers were then prepared at different deposition temperatures of homo-buffer layer ($T_{\text{homo-buffer}}$ = 30, 100 and 500 °C) and different deposition oxygen pressures of homo-buffer layer ($P_{\text{homo-buffer}}$ = 0, 40 and 200 mTorr). AZO films were prepared under the above optimized deposition condition ($A_1B_3C_2D_2$). In addition, the AZO film without buffer layer (the bare AZO film) was deposited for comparison.

3. Result and discussion

3.1. The influence of $T_{\text{homo-buffer}}$ on properties of AZO film

Fig. 1(a) displays the XRD patterns of AZO films with homo-buffer layer and bare AZO film. The films exhibit obvious (0 0 2)

Table 2
The orthogonal array and the relative data of range analysis.

Number	Factors				Evaluation index (%)
	Oxygen pressure	Laser energy	Substrate temperature	Deposition time	
1	A_1	B_1	C_1	D_1	T_1 = 83.198
2	A_1	B_2	C_2	D_2	T_2 = 85.052
3	A_1	B_3	C_3	D_3	T_3 = 81.940
4	A_2	B_1	C_2	D_3	T_4 = 82.463
5	A_2	B_2	C_3	D_1	T_5 = 77.819
6	A_2	B_3	C_1	D_2	T_6 = 82.779
7	A_3	B_1	C_3	D_2	T_7 = 79.845
8	A_3	B_2	C_1	D_3	T_8 = 83.117
9	A_3	B_3	C_2	D_1	T_9 = 82.221
$\bar{k}_{1j}$	83.397	81.835	83.031	81.079	
$\bar{k}_{2j}$	81.020	81.996	83.245	82.559	
$\bar{k}_{3j}$	81.728	82.313	79.868	82.507	
The optimal level	A_1	B_3	C_2	D_2	
R_j	2.377	0.478	3.377	1.480	

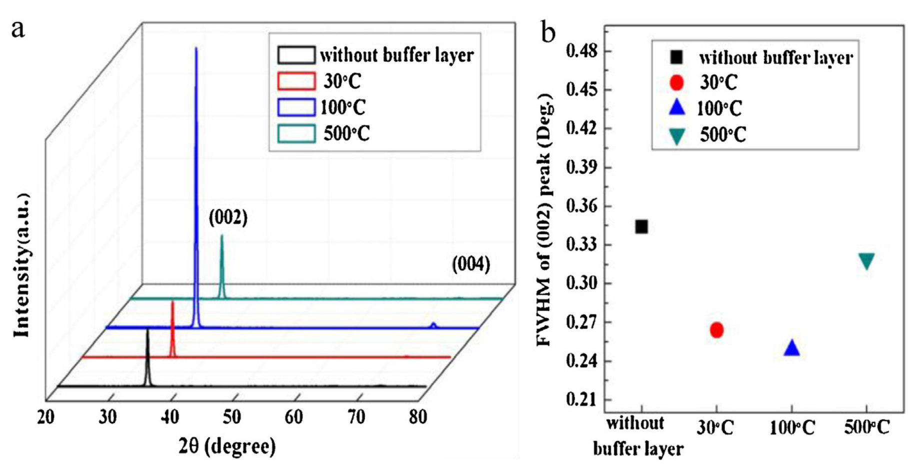

Fig. 1. (a) XRD patterns of the bare AZO films and AZO films with homo-buffer layer deposited under various $T_{\text{homo-buffer}}$ and (b) corresponding FWHM as a function of $T_{\text{homo-buffer}}$.