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Structure, optical constants and non-linear properties of high quality AZO nano-scale thin films

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ABSTRACT

Al-doped ZnO thin films of thickness ≅60 nm were prepared by RF reactive magnetron sputtering. All optimum conditions were fixed through the depositions expect the power of the DC for Al-dopants. The structure of the as-grown samples measured by XRD showed that the films have a hexagonal structure with a space group *p*63*m*. The SEM micrographs illustrated uniform and/or non-uniform spherical shape for pure ZnO and Al-doped ZnO, while the sizes ranged from 18 to 35 nm. The linear refractive indices (n) were calculated on the basis of Fresnel's equations. The refractive index pounced peaks changed with changing the Al-dopnats. Based on the spectroscopic data, the linear and non-linear parameters such as the linear optical susceptibility $\chi^{(1)}$, non-linear refractive index (n_2) or non-linear optical susceptibility $\chi^{(3)}$ were calculated and analyzed from spectroscopic analysis. The Al-doped ZnO is a good candidate for new generation of electronic and optoelectronic devices.

1. Introduction

Transparent conducting oxides (TCOs) are the promising oxides materials for new generation of electronic and optoelectronic devices [1–5], where its applications are widely used in optical and magnetic memory devices, transducer, solar cells, flexible displays, flat panels, liquid crystal displays, blue or ultra-violet (UV) LEDs, gas sensor, laser diode, organic light-emitting diodes (OLED), piezo-electric device, surface-acoustic-wave, chemical sensors, thin-film transistors, thin-film solar cells and acts as a back contact for new inverted devices. . .etc. [1–5].

The best and abundant TCO materials are Al-doped ZnO (AZO) due to its low toxicity, highly transparent films, high stable metal oxides and its low resistivity (highly conductive) [6]. AZO films can deposited in different substrates by different methods such as: sol–gel method, pulsed laser depiction (PLD), RF magnetron sputtering, Atomic layer deposition. . .etc. [7–9]. RF magnetron sputtering is a unique technique to prepare high quality AZO films with high reproducibility and/or reliability, large scale area, controllable rate of deposition, high stability films and favorable for flexible substrates, smooth, highly-adherent films and efficient technological transfer to industry [10,11].

In this work, pure and Al-doped ZnO nanostructure thin films are grown successfully on glass substrates at room temperature (RT). XRD and FE-SEM are used to analyze the structure and surface morphology of nanostructure AZO films. Optical constants were calculated on the basis of Fresnel's equations. Optical band gap was calculated on the basis of Tauc's model. Our goals is to establish approximation method to calculate the non-linear optics of AZO by spectroscopic method. The analyzing of the linear refractive index (n) and linear optical susceptibility $\chi^{(1)}$ has been used for nonlinear parameter calculations such as: non-linear refractive index (n_2), and non-linear optical susceptibility $\chi^{(3)}$.

2. Experimental technique

2.1. Preparation

Co-sputtering system has the advantage of allowing an easy tuning and control of the percentage of Al in the AZO film. AZO thin films were grown on a glass substrate and silicon wafer using radio frequency (DC/RF) magnetron sputtering (DC/RF Magnetron Sputter System, Syskey Technologies, Taiwan). The glass slides

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Table 1
The relation between the Al DC sputtering power and wt% for AZO thin films.

Al-DC power (W)	0	20	30	40	50	60
Al-concentrations in (wt%)	0	3.4	4.3	4.5	4.8	5.3

were thoroughly cleaned with acetone and dried with pressurized nitrogen before deposition. Highly purity targets for both ZnO (3 × 0.6 in.) and Al metal (3 × 0.6 in.) were used for deposition of AZO. For pure ZnO, 200 W power was applied by RF inside sputtering chamber, while Dc power ranging 20–60 W was used for Al. The base pressure in the chamber adjusted to 1×10^{-6} Torr before introducing Argon gas, while operating pressure of 5×10^{-3} Torr maintained with substrate rotation at 15 rpm and deposition time 600 s. The relation between the Al DC sputtering power and wt% for Al doped ZnO is shown in Table 1, where the wt% of Al increased with increases the Al RF sputtering power.

2.2. Devices

X-ray diffractometer (Ultima-IV; Rigaku, Japan) with CuK_α radiation $\lambda = 1.5418$ Å wavelength having 40 kV accelerating voltage and 30 mA current. UV–visible absorption and transmittance spectra were recorded using the UV–visible spectrophotometer, Perkin Elmer, Lambda 750. The reflectance was calculate following equation ($T + R + abs = 1$). The morphology was studied by a field emission scanning electron microscope (FESEM) (JSM-7500 F; JEOL-Japan) using parallel beam geometry and a multi-purpose thin film attachment.

3. Results and discussions

3.1. Structure properties

Fig. 1 shown the XRD pattern of pure and Al doped ZnO with different concentrations of Al(3.4–5.3%). All films exhibit only two position peaks at $2\theta^\circ$ which equal 34.178 and 62.5 with small changes due to the different Al concentrations. The diffractions angle at 34.178 represents [0 0 2] plane while the angle at 62.5 corresponding to [1 0 3] plane according to JCDPS card No. (00-003-0888). XRD supports the single ZnO Phase for the as-deposited samples i.e. no any secondary phase corresponding to the Al_2O_3 or other intermediate compound. Also, we can confirmed that there is no metallic zinc or aluminum characteristic peaks were observed

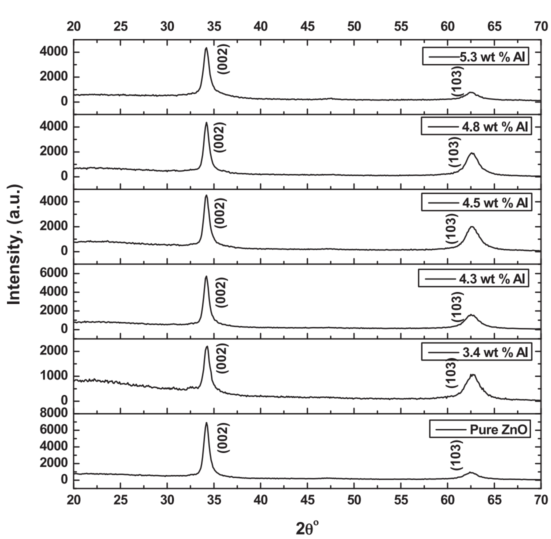

Fig. 1. XRD patterns of for nanostructure AZO thin films grown on glass substrates at room temperature for.

in the studied AZO films. The behavior of these films is reported by other authors [12]. The crystallite size of pure and Al-doped ZnO can be calculated from the well known Scherrer's equation based on the full width at half maximum (FWHM) as follows [13–15]:

$$D = \frac{0.9\lambda}{\beta \cos\theta} \quad (1)$$

where β is the full-width at the half maximum. The calculated crystallite size of pure and Al-doped ZnO is given in Table 2. The value of the crystallite size changes irregular (unsystematically) with increasing the Al-dopants within the range from 11.389 to 13.073 nm for [0 0 2] plane and from 4.969 to 5.373 nm) for [1 0 3] plane. Both dislocation density δ and the strain ε were calculated to study the structure properties of pure and Al-doped ZnO from the following relations [16–19]:

$$\delta = \frac{n}{D^2}, \quad (2)$$

and

$$\varepsilon = \frac{\beta \cos\theta}{4}, \quad (3)$$

where $n = 1$ at the minimum dislocation density. The defect dislocation density can be defined as the length of the dislocation line per unit volume [16]. The values of δ are in the range 7.709×10^{-3}–4.049×10^{-2} (nm)$^{-2}$ for the studied pure and Al-doped ZnO planes. The dislocation density describes the quality of the AZO films and so, the lower values of δ, the high quality of the crystallized films. The obtained results are in good agreement with the reported data for ZnO thin films [20]. The calculated lattice strain for AZO films are in the range 3.044×10^{-3}–6.975×10^{-3} (nm)$^{-2}$.

FE-SEM measurement of the surface morphology of pure and Al-doped ZnO with different wt% of Al-dopants is sown in Fig. 2. From the analysis of FE-SEM micrographs, both pure and Al-doped ZnO have a uniform and/or non-uniform spherical shape, with grain size ranged from 18 to 35 nm. There are correlations between XRD and SEM measurements which support the nano-structure of the as-prepared pure and Al-doped ZnO nano-structure thin films.

3.2. Linear and non-linear optics of AZO thin films

The wavelength dependence of both transmittance $T(\lambda)$ and reflectance $R(\lambda)$ are shown in Fig. 3. All optical measurements are done at normal light for all samples. The transmittance of the studied samples is very high at the higher wavelengths, in which the light passes through the sample without any absorption (transparent region i.e. no light absorbed with $T(\lambda) + R(\lambda) \approx 1$) [21]. The average transmittance values of samples in all light regions in the range 80–90% exhibit the absorption edge. The films have shown a decrease in its value for lower wavelengths manifest with the formation of valleys and peaks located in the range 1250 to 406 nm. The absorption edges (i.e. the sharp transmission edges) are appeared in UV region and corresponding to electronic transitions from valance band to conduction band [22]. The relation between the reflectance $R(\lambda)$ and the linear refractive index (n) can be described by the Fresnel's formula as follows [23–25]:

$$R = \frac{(n-1)^2 + k^2}{(n+1)^2 + k^2}, \quad (4)$$

Based on the algebraic solution, the refractive index can be expressed by [23–25]:

$$n = \frac{(I+R)}{(1-R)} + \sqrt{\frac{4R}{(1-R)^2} - k^2}, \quad (5)$$