Structural, Surface Morphology and Optical Properties of Pulsed Laser Deposited ZnO Thin Films at Various Laser Pulse Energies

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Abstract. ZnO thin films have been deposited on ITO substrates by pulsed laser deposition at various laser pulse energies. The crystalline structure, surface morphology and optical properties of ZnO thin films were characterized by X-ray diffraction (XRD), atomic force microscope (AFM) and UV-Visible measurements. The results showed that ZnO thin films grew along with (002) preferential orientation and had optimal crystalline quality when laser pulse energy kept in 180mJ·P^{-1}. In addition, the films had smooth surface and compacted structure. Laser pulse energy had little influence on the transmittance of ZnO thin films in visible light region and the transmittance of all the ZnO thin films is above 80%. It also can be found that the film thickness and optical band gap increased with the increase of laser pulse energy from 150mJ·P^{-1} to 200mJ·P^{-1}.

Introduction

ZnO is a semiconductor material with a direct wide band gap (~3.37eV), and have a wide range of promising applications for solar cells, light emitting diodes, transparent electrodes and UV photodetector, due to its outstanding optical properties, high exciton binding energy, excellent thermal stability and low cost [1]. Although ZnO has lots of excellent properties, the abundance of defects in ZnO is an impediment for wide application [2]. Nowadays, there are many different methods to prepare ZnO, such as magnetron sputtering [3], sol-gel coating [4], chemical vapor deposition (CVD) [5], pulsed laser deposition (PLD) [6] and spray pyrolysis [7].To achieve high quality ZnO thin films, we use pulsed laser deposition which have advantage of simple operation, low substrate temperature and good consistency of film and target composition.

In this paper, ZnO thin films were deposited on ITO substrates by pulsed laser deposition at various laser pulse energies. The effects of laser pulse energy on structural, surface morphology and optical properties of ZnO thin films were investigated.

Experimental

ZnO thin film was deposited on ITO substrate by LMBE-450 pulsed laser deposition system. Prior to the deposition, the ITO substrates were ultrasonically cleaned sequentially in acetone， ethyl alcohol and de-ionized water for 5 min, respectively. A pure ceramic ZnO target (99.99%) was used to deposit ZnO thin films, and the substrate was placed 4 cm away from the ZnO target. The base pressure was pumped down to 10^{-5} Pa, and the substrate was heated to 473K. During the growth of ZnO thin film, oxygen gas (99.999% purity) was introduced into the deposition chamber

to reach a working pressure of 0.8 Pa. Laser pulse energy was varied from 150mJ·P^{-1} to 200mJ·P^{-1} (KrF excimer laser beam, l ¼ 248 nm and n ¼ 5 Hz). All depositions were carried out for 60min.

The crystalline phase analysis of ZnO thin films was identified by X-ray diffraction (Rigaku D/MAX 2500) using Cu Kα radiation (λ=0.154nm), the source being operated at 40kV and 40mA. The surface morphology of films was studied by Agilent 5500 atomic force microscope. The optical transmission was measured by PerkinElmer Lambda 950 UV-Vis-NIR spectrophotometer along with 150mm integrating sphere.

Results and discussion

Fig. 1 shows XRD patterns of ITO substrate and ZnO thin films deposited at various laser pulse energies. The absence of ZnO thin films peaks at 2θ in the XRD pattern were 34.8° and 63.2°, corresponding to the Miller indexes of the reflecting planes of hexagonal wurtzite ZnO (002) and (103) (JCPDS no.36-1451). ZnO thin films show a (002) preferential orientation and a weak (103) orientation. As the laser pulse energy increases from 150mJ·P^{-1} to 180mJ·P^{-1}, the relative intensity of the peak in (002) plane increase for ZnO thin films while it decrease for ZnO thin films on elevating laser pulse energy from 150mJ·P^{-1} to 180mJ·P^{-1}.

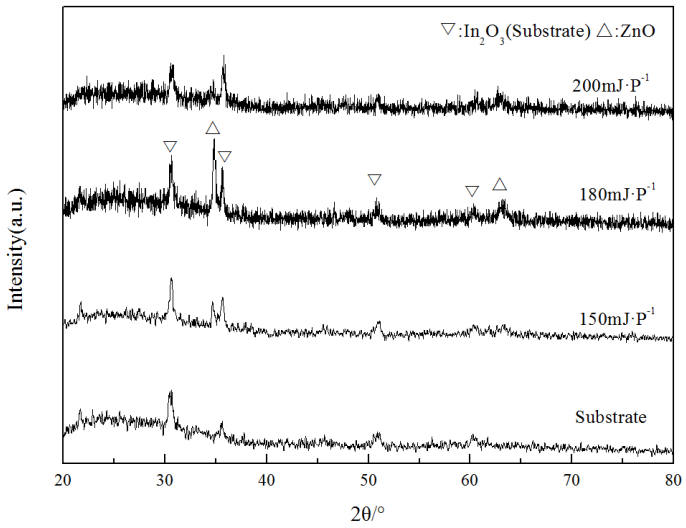

Fig. 1 XRD patterns of ITO substrate and ZnO thin films deposited at various laser pulse energies

AFM patterns of ZnO thin films deposited at various laser pulse energies are illustrated in Fig. 2, revealing that a grain size is 80~160nm. The superficial roughness was given in Table 1.While ZnO deposited at 150mJ·P^{-1} to 180mJ·P^{-1}, the films has smooth surface and compacted structure. However, ZnO thin films have lots of voids between relatively isolated grains at 200 mJ·P^{-1}, and the superficial roughness is much higher than ZnO thin films deposited at 150mJ·P^{-1} to 180mJ·P^{-1}.

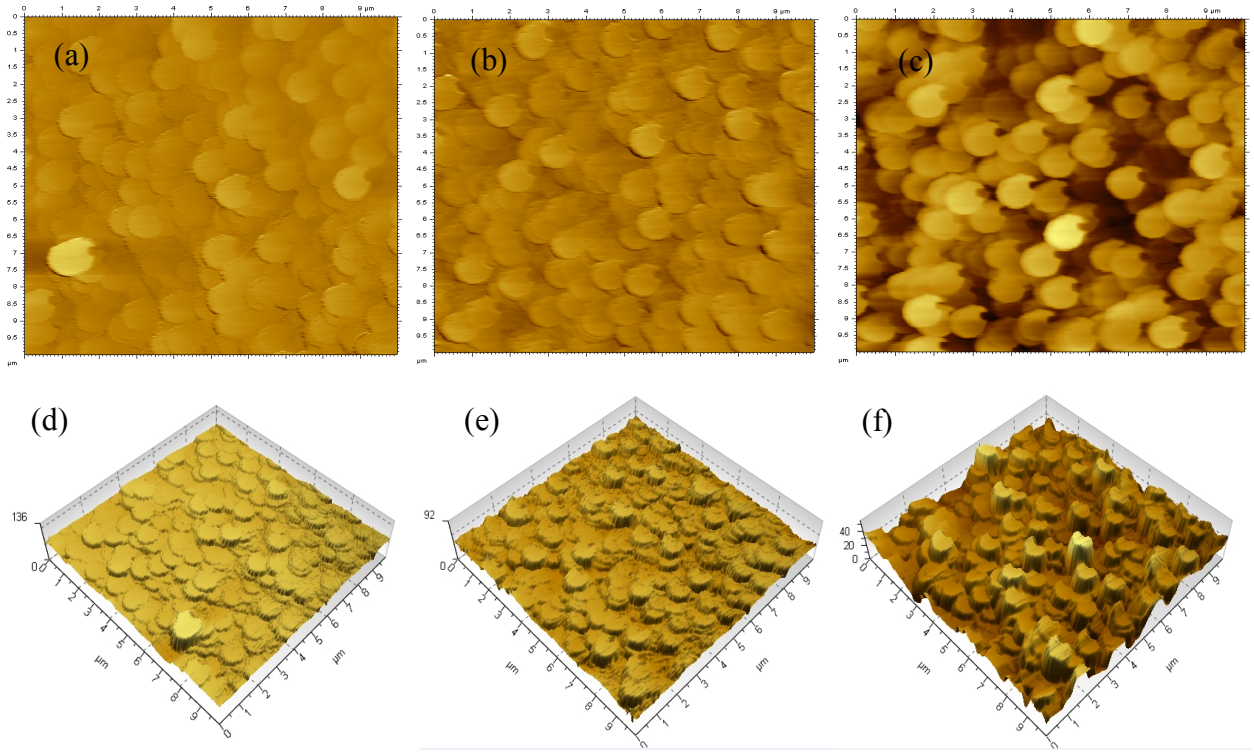

Fig. 2 AFM patterns of ZnO thin films deposited at various laser pulse energies (10μm×10μm) (a) and (d): 150mJ·P^{-1} (b) and (e): 180mJ·P^{-1} (c) and (f): 200mJ·P^{-1}

Table.1 Superficial roughness of ZnO thin films deposited under various laser pulse energies

Laser pulse energy (mJ·P^{-1})	Arithmetic mean roughness (nm)	Root mean square roughness (nm)
150	3.7	4.9
180	3.6	4.7
200	6.2	7.8

Fig. 3(a) shows the transmittance spectra of ZnO thin films deposited under various laser pulse energies. Laser pulse energies had little influence on the transmittance of ZnO thin films in the visible range and the transmittance of all the ZnO thin films is above 80%. As the laser pulse energy increases from 150mJ·P^{-1} to 180mJ·P^{-1}, the absorption edge shifts to shorter wavelength.

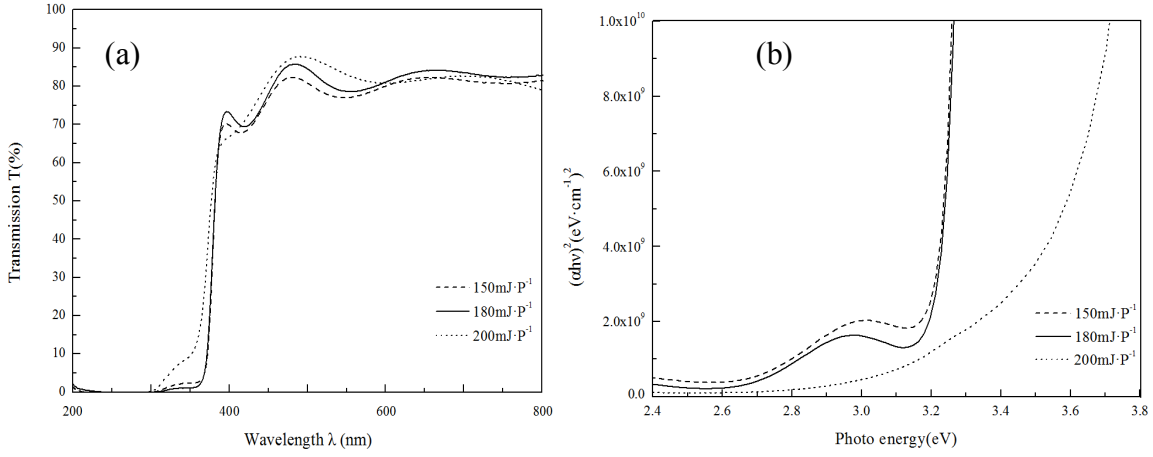

Fig.3 (a) Transmittance spectra and (b) Plots of $(\alpha h\nu)^2$ as function of photo energy of ZnO thin films deposited at various laser pulse energies

Fig.3 (b) shows Plots of $(\alpha h\nu)^2$ as function of hν of ZnO thin films deposited at various laser pulse energies. The films thickness and the optical band gap (E_g) of ZnO thin films, calculated by optical transmittance data, were reported in Table 2 [8]. The calculated optical band gap of the films varies between 3.19eV and 3.32eV. It can be observed that the optical band gap and the film thickness increase with the increase of laser pulse energy from 150mJ·P^{-1} to 200mJ·P^{-1}.

Table 2 Film thickness and optical bandgap energy (E_g) of ZnO thin films deposited at various laser pulse energies

Laser pulse energy（mJ·P^{-1}）	Film thickness（nm）	E_g (eV)
150	259	3.19
180	268	3.21
200	329	3.33

Conclusions

In this study，the change of structural, surface morphology and optical properties of ZnO thin films deposited at laser pulse energy have been explored. Experimental results show that the deposited films are hexagonal wurtzite structure which has a (002) preferred growth orientation. ZnO thin films had better crystalline quality and smooth surface when laser pulse energy was 180mJ·P^{-1}. Films deposited at higher laser pulse energy show increased the optical band gap value of ZnO thin films from 3.19 eV to 3.32eV. Laser pulse energy had little influence on the transmittance of ZnO thin films in visible light region and all the ZnO thin films show above 80% transmittance in visible range.

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