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ScienceDirect
Physica B 396 (2007) 95–101
PHYSICA B
www.elsevier.com/locate/physb

Structural and optical properties of ZnO thin films on glass substrate grown by laser-ablating Zn target in oxygen atmosphere

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Received 4 February 2007; received in revised form 26 February 2007; accepted 14 March 2007

Abstract

C-axis-orientated ZnO thin films were prepared on glass substrates by pulsed-laser deposition (PLD) technique in an oxygen-reactive atmosphere, using a metallic Zn target. The effects of growth condition such as laser energy and substrate temperature on the structural and optical properties of ZnO films had been investigated by X-ray diffraction (XRD), scanning electron microscopy (SEM), transmission spectra and room-temperature (RT) photoluminescence (PL) measurements. The results showed that the thickness, crystallite size, and compactness of ZnO films increased with the laser energy and substrate temperature. Both the absorption edges and the UV emission peaks of the films exhibited redshift, and UV emission intensity gradually increased as the laser energy and substrate temperature increased. From these results, it was concluded that crystalline quality of ZnO films was improved with increasing laser energy and substrate temperature.

PACS: 81.15.Fg; 81.05.Dz; 68.55.Jk; 78.66.Hf

Keywords: ZnO; Pulsed-laser deposition (PLD); Optical properties; Deposition process

1. Introduction

Zinc oxide (ZnO) is an interesting wide band gap (~3.3 eV) II–VI semiconductor with potential application in optical-electrical field, such as ultraviolet (UV) light-emitting diodes, UV photodetectors, transparent thin-film transistors, surface acoustic wave (SAW) filter, and gas sensors [1–5]. In recent years, various techniques such as sol–gel, spray pyrolysis technique, electrochemical deposition, metalorganic chemical vapor deposition (MOCVD), molecular beam epitaxy (MBE), vacuum arc technique, radio-frequency (RF), magnetron sputtering, and pulsed-laser deposition (PLD) had been developed to prepare ZnO thin films [6–13]. Using PLD technique, ZnO thin films can be formed at a relatively high oxygen partial pressure and lower temperature due to higher energy of the ablated particles in the laser-produced plume [14]. Up to date, much works concerned with pulsed-laser deposited ZnO films from ZnO ceramic target were reported [15]. Considering that the metallic target is denser, purer, and easier to fabricate in comparison with the normally used ZnO ceramic target, some researchers use Zn target to prepare the ZnO films by the pulsed-laser reactive ablation technique [16–20]. Generally, the excimer laser with shorter wavelength has been widely used to grow ZnO thin films although ZnO thin films with relatively high quality are also obtained by the laser with longer wavelength [21,22]. In this paper, ZnO thin films deposited on glass substrate were obtained by using an excimer laser-ablating high-purity Zn target in oxygen-reactive atmosphere. The influences of process parameters including laser energy

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0921-4526/$ - see front matter
doi:10.1016/j.physb.2007.03.018

and substrate temperature on the structural and optical properties of the films were investigated.

2. Experimental procedure

ZnO thin films were deposited onto glass substrates in a PLD system. The target was a metallic Zn disk (99.9% purity) with 2.5 cm in diameter and 0.5 cm in thickness. Glass sheets, which were used as the substrates for ZnO deposition, were cleaned in an ultrasonic bath with acetone for 10 min before being loaded into the chamber. The substrates were placed parallel to the target surface at a distance of 5 cm. A pulsed excimer laser (KrF; $\lambda = 248$ nm, COMPex205, Lambda Physik) was used with pulse duration of 25 ns and repetition rate of 5 Hz. The laser beam was focused through a 50 cm focal lens onto a rotating target at a 45° angle of incidence. The deposition chamber was initially evacuated to 3×10^{-3} Pa, and the oxygen pressure and deposition time were fixed at 100 Pa and 10 min, respectively. At a fixed substrate temperature of 350 °C, four ZnO films were prepared with laser energy of 100, 175, 250, and 325 mJ/pulse, respectively. Other three ZnO films were prepared under different substrate temperatures (RT, 200 and 500 °C) at a fixed laser energy of 250 mJ/pulse.

An X-ray diffraction apparatus (XRD, BDX3200, Perking University, China) with Cu $K_{\alpha 1}$ incident radiation was used to identify the phase structure of the grown films. The surface morphology of the films was studied by using a field emission scanning electron microscope (FE-SEM, Sirion, FEI). The film thickness was measured by a nanostep instrument (Form Talysurf S4C, Taylor Hobson). The optical transmittance properties of ZnO films were measured by using an UV–vis–Nir spectrometer (Cary 5000, Varian). Photoluminescence (PL) spectra of films were measured by using a He–Cd laser at 325 nm.

3. Experimental results and discussion

3.1. Structural properties

Fig. 1(a) shows the XRD patterns of the films grown at different laser energies. At all laser energies, only one diffraction peak of ZnO phase is found in their XRD patterns. The intensity of diffraction peak for the films grown at 100 mJ/pulse is very weak, but it increases with increasing laser energy. The XRD patterns of the films grown at different substrate temperatures are shown in Fig. 1(b). No diffraction peak is observed in the films grown at the substrate temperature of RT. When the substrate temperature increases to 200 °C, only one diffraction peak of the ZnO phase is observed, and its intensity increases as the substrate temperature further increases. It is worth noting that the diffraction peak of the ZnO phase is identified as (0 0 2), which indicates that the ZnO films prepared by reactive PLD show a good *c*-axis orientation perpendicular to the substrate.

As shown in Fig. 2(a), the full width at half maximum (FWHM) of (0 0 2) diffraction peak decreases from 0.4° to 0.29° with increasing laser energy, which revels that the crystalline quality of films is improved. Obviously, crystallinity of the films is promoted by increasing the substrate temperature from RT to 200 °C, as mentioned above. However, it is seen that the change of FWHM for (0 0 2) diffraction peaks is not remarkable when substrate temperature increases from 200 to 500 °C, as shown the Fig. 2(b). By applying the Scherrer equation to the FWHM of the (0 0 2) peaks [23], average crystallite size of the ZnO films grown at different laser energies are 20, 25, 27, and 28 nm, respectively, and that of the films grown at 200–500 °C are about 27 nm.

It is seen form Fig. 2 that the diffraction angle increases as the laser energy increases from 100 to 250 mJ/pulse, then it begins to decrease when the laser energy further increases to 325 mJ/pulse; the maximal diffraction angle is observed

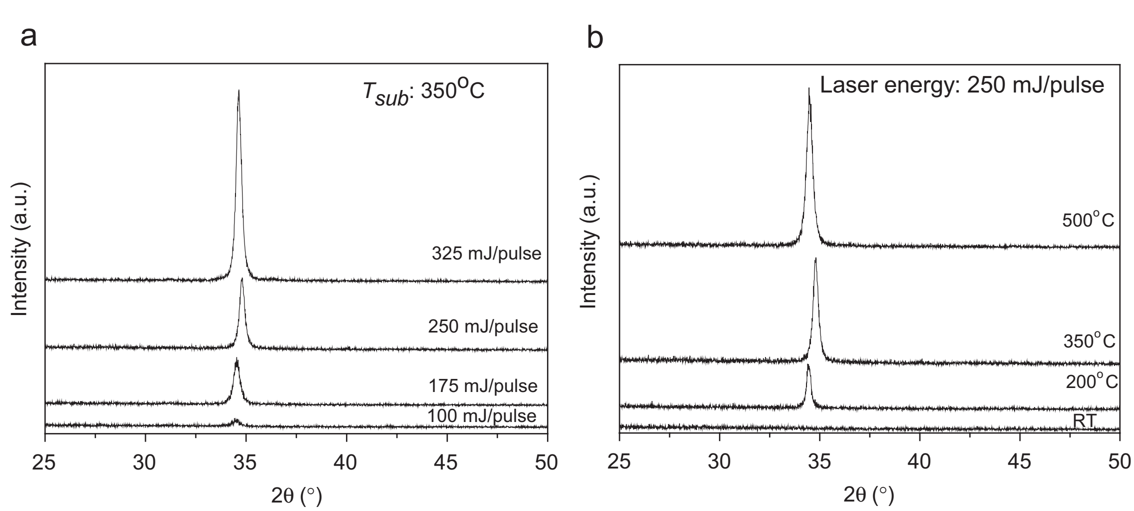

Fig. 1. XRD spectra of ZnO films deposited on glass substrates at various laser energies (a) and substrate temperatures (b).