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Microstructure and optical properties of polycrystalline ZnO films sputtered under different oxygen flow rates

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ABSTRACT

Polycrystalline ZnO films were fabricated using rf magnetron sputtering under different oxygen flow rates (P_O). The surface morphology of the films can be affected by changing P_O, and the average surface roughness decreases with the increasing P_O. The increasing P_O can improve the grain growth with (0 0 2) orientation. Typical ZnO infrared vibration bands have been observed at 408 and 513 cm^{-1}, and the full width at half maximum of the infrared peak decreases with the increase of P_O due to the improved crystallinity. The optical band gap (E_g) of the polycrystalline ZnO films increases from 3.22 eV at $P_O = 0$ sccm to 3.25 eV at $P_O = 10$ sccm because the defects decrease with the increasing P_O. The photoluminescence peaks in the region of 2.4–2.7 eV are from the transition between conduction band edge and oxide antisite defects (O_{Zn}), and also be influenced by oxygen vacancies (V_O).

1. Introduction

ZnO with wide band gap (3.37 eV) and large binding energy (60 meV) has attracted considerable attention from both fundamental and application points of view, such as solar cells, blue and ultraviolet light emitting devices [1–4]. Furthermore, ZnO nanostructures can also be used in various applications such as nanowire, nanolasers, biosensors and field emission devices [5]. Piezoelectric properties of ZnO are being explored for fabrication of various pressure transducers, acoustic and optoacoustic devices [6]. Zn vacancies induced magnetic order in the pure ZnO films has been reported by Khalid et al. [7]. With the development of material science, ZnO will offer more applications in the associated fields. Nano-size polycrystalline and epitaxial ZnO films have been extensively fabricated by using several methods including pulse laser deposition [8,9], molecular beam epitaxy [10–12], magnetron sputtering [13,14], oxidation of metallic zinc films [15], sol–gel method [16], etc. The surface morphology, microstructure, film growth, electrical and optical properties of ZnO films can be affected by the experimental conditions, such as substrates, fabrication methods, oxygen flow rate, and element doping [17–21], which can be ascribed to the change of the defects in the films. It is well known that the sputtering technology is an effective and the most widespread method for fabricating thin films in industry nowadays, and the oxygen flow rate can affect the intrinsic defects in the ZnO films during the film deposition significantly. In this paper, polycrystalline ZnO films were fabricated using rf magnetron sputtering from ZnO target under different oxygen flow rates. Morphology, microstructure and optical properties of the polycrystalline ZnO films are investigated systematically using atomic force microscopy, X-ray diffraction, infrared spectroscopy, UV–vis transmission spectroscopy, and photoluminescence spectroscopy.

2. Experimental details

Polycrystalline ZnO films were fabricated using rf magnetron sputtering method at room temperature from ZnO target in Ar and O_2 gas mixture on glass and Si (1 0 0) wafers with natural oxide layer. The base pressure of the chamber reached 6.0×10^{-6} Pa or better. During the film deposition, the total pressure of sputtering gas was kept at 1.0 Pa (100 sccm) and the O_2 flow rate (P_O) was changed from 0 to 10 sccm. The film thickness was about 300 nm determined using a Dektak 6 M surface profiler. The morphology, structure, and chemical states of the films were characterized using atomic force microscopy (AFM), X-ray diffraction (XRD), and Fourier transform infrared spectroscopy (FTIR, Brucker 8 V). The UV–vis transmission spectra were measured using the UVPC spectrophotometer system (Shimadz

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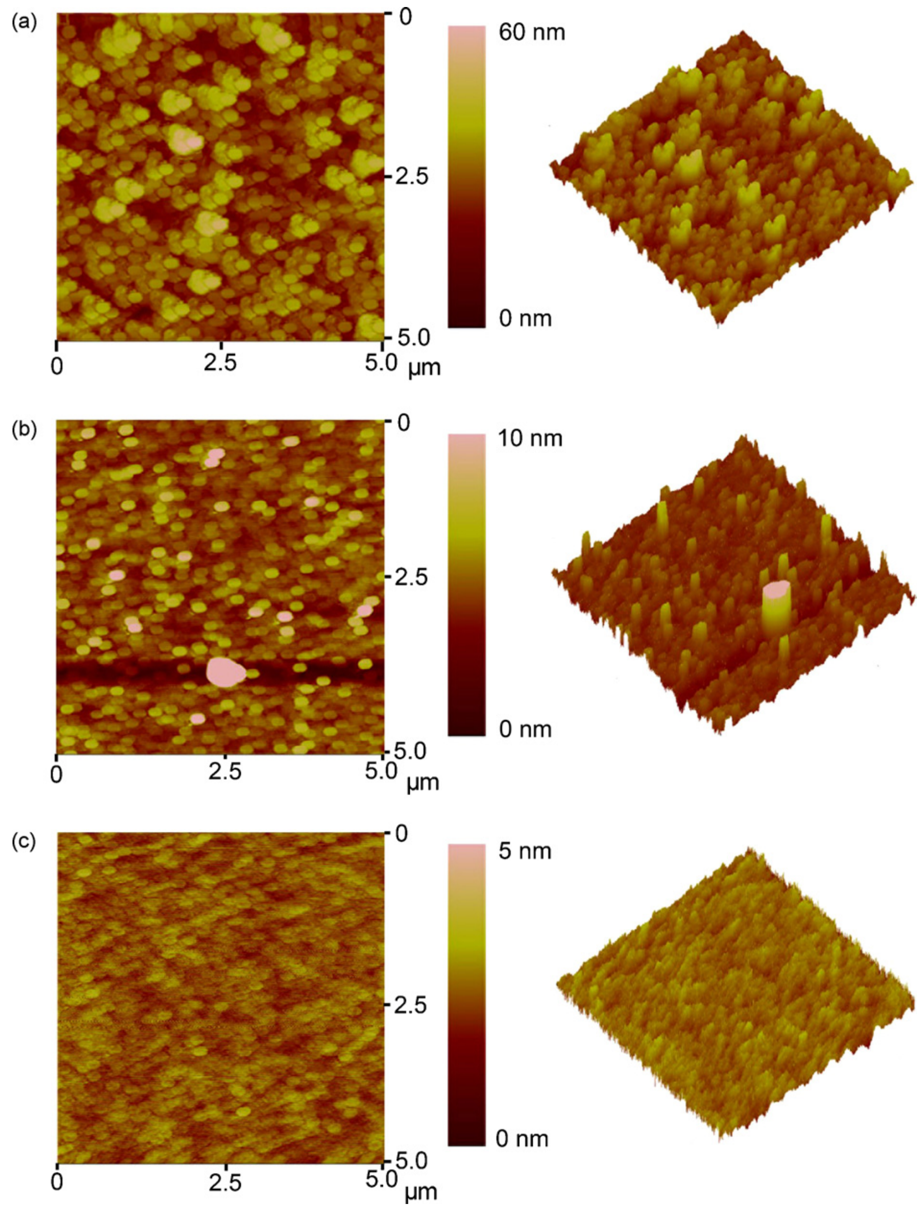

Fig. 1. AFM images of the polycrystalline ZnO films fabricated at different P_O, (a) $P_O = 0$ sccm, (b) $P_O = 2$ sccm, and (c) $P_O = 6$ sccm.

3101 PC). Photoluminescence spectra (PL) were used to study luminescence properties of the films recorded by a Hitachi F-4500 fluorescence spectrophotometer with Xenon lamp as the excitation light source at room temperature.

3. Results and discussion

3.1. Morphology and microstructure

Fig. 1 shows the AFM images of the polycrystalline ZnO films fabricated at different P_O. From this figure, one can see that at $P_O = 0$ sccm, the film surface is composed of uniform clusters with a size of 500 nm, and the clusters are composed of several uniform 200-nm islands. For observing the surface morphology of the film clearly, the large-scale AFM image of the film fabricated at $P_O = 0$ sccm is given in Fig. 2. From Fig. 2, one can see that the islands on the film surface are so uniform, and the distance between the large clusters is similar, but the film surface is not so smooth. At $P_O = 2$ sccm, there are some rods with the diameter of about 200 nm on the film surface, and the rods are separated with the large distance of 300–2000 nm and arbitrary distribution. The film surface looks more smooth, and the longitudinal height is in the scale of 10 nm. As P_O increases to 6 sccm, no obvious islands can be observed on the film surface, and the film surface is so smooth in the scale of 5 nm. The average surface roughness R_a is defined as the arithmetic average deviation from the mean line within the assessment length L

$$R_a = \frac{1}{L}\int_{x=0}^{x=L} |y|dx, \quad (1)$$

where x is the displacement along the lateral scan direction and y is the vertical fluctuation. The average surface roughness of the films decreases from 5.155 nm at $P_O = 0$ sccm to 0.419 nm at $P_O = 6$ sccm. The surface morphology should be determined by crystal growth mechanisms and the diffusion behavior during the film deposition. In the present samples, the change of the oxygen flow rate can affect the crystal growth and diffusion of the atoms/ions deposited on the substrates, so the surface morphology of the films fabricated at different P_O is different.