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Structural Properties of Sputter-Deposited ZnO Thin Films Depending on the Substrate Materials

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Abstract. We have demonstrated the growth of ZnO thin films with c-axis orientation at room temperature on various substrates such as Si(100), SiO_2, and sapphire by the r.f. magnetron sputtering method. X-ray diffraction (XRD) and scanning electron microscopy altogether indicated that the larger grain size and the higher crystallinity were attained when the ZnO films were deposited on sapphire substrates, compared to the films on Si or SiO_2 substrates. The c-axis lattice constant decreased by thermal annealing for the ZnO films deposited on Si or SiO_2 substrates, while increased by the thermal annealing for the ZnO films grown on sapphire substrates.

Introduction

Zinc oxide (ZnO) thin films received much attention due to its unique piezoelectric and piezooptic properties made suitable for acoustic devices [1], low loss optical waveguides [2], optoelectronic devices [3, 4], and gas sensors [5]. Additionally, ZnO has been developed as a promising alternative to transparent conducting indium tin oxide and tin oxide coating due to its low cost and nontoxicity [6]. ZnO have been prepared by various physical vapor deposition techniques such as evaporation, r.f./d.c. sputtering [7,8], ion beam sputtering, atomic layer epitaxy etc., as well as a plethora of chemical methods [9]. Among these methods, r.f. magnetron sputtering has gained importance due to its potential for low-cost and low temperature process. Although many researchers have grown ZnO films, the effects of substrate material on the structural characteristics of ZnO thin films have not been systematically investigated. In this paper, we have deposited the c-axis oriented ZnO films by the r.f. magnetron sputtering method. We have studied the dependence of the structural properties on the substrate material, i.e. silicon (Si), SiO_2, and sapphire.

Experiments

A schematic description of the r.f. sputtering system is previously reported [10]. ZnO films were deposited by the r.f. magnetron sputtering system using a ZnO (99.99 % purity) target with a diameter and a thickness of 75 mm and 6 mm, respectively. The sputtering was carried out in a 30 sccm Ar (99.99 % purity) gas atmosphere by supplying 80-250 W r.f. power. The magnetron is capacitively powered with 13.56 MHz radio-frequency. In ordet to exclude the thickness effect, we have fixed the ZnO film thickness to about 500-600 nm.

The ZnO thin films were sputtered on Si, SiO_2, and sapphire substrates. The Si substrate was p-type silicon with (001) orientation and the resistivity was 1-30 ohm-cm. The Si substrate was cleaned in acetone for 10 min, HF (20:1) for 10 seconds and then rinsed by deionized water for 1 minute before loading into the sputtering system. The (0001)-oriented sapphire and the SiO_2 substrates were cleaned in acetone for 10 min. The SiO_2 substrates were produced by thermally growing a 60 nm-thick SiO_2 layer on the Si substrate.

The chamber was down to 6 x 10^{-6} torr using a turbo-molecular pump before introducing the Ar sputtering gas into the chamber. The ZnO film was grown at room temperature at a pressure of 5.0 x 10^{-2} torr. The thermal annealing was performed in a furnace using quartz tube reactor at a temperature of 900 ^{o}C in oxygen gas flow of 3 standard liter per minute for 10 minutes. The structural properties of the films were analyzed by X-ray diffraction (XRD) using CuKα1 radiation (λ = 0.154056 nm) and by scanning electron microscopy (SEM) (Hitachi S-4200).

Results and discussion

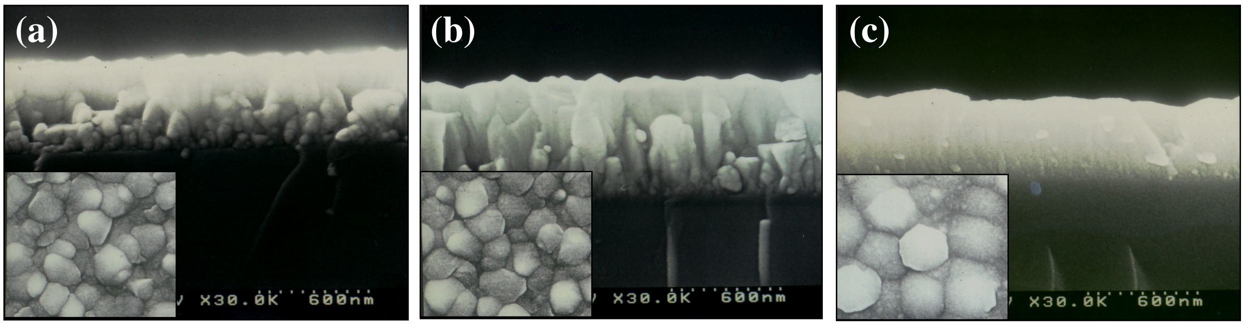

Fig. 1. Cross-sectional (Inset: plan-view) SEM images of ZnO films grown on (a) Si, (b) SiO_2, and (c) sapphire substrate.

We have compared the cross-sectional SEM images of ZnO films grown on Si, SiO_2, and sapphire substrates in Fig. 1, in which the RF power was set to 150 W for equal growth time. The images indicate that the crystallinity of the film on sapphire substrate is higher than that on Si or SiO_2 substrate. We surmise that relatively little lattice mismatch between ZnO and sapphire substrates helps to grow the higher crystalline films. The plan-view SEM images reveal that the average grain size on top of ZnO films deposited on sapphire substrates is larger than those deposted on Si or SiO_2 substates.

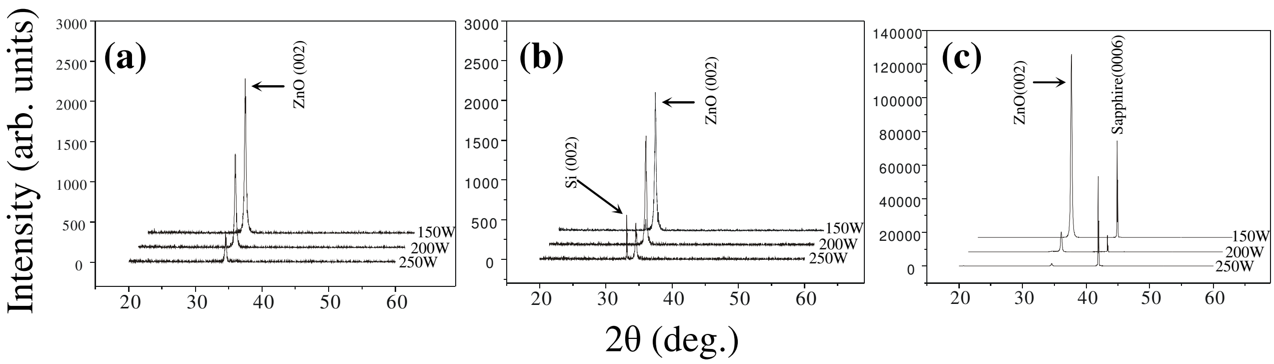

Fig. 2. The relative intensity of (002) diffraction peak of ZnO films deposited on (a) Si, (b) SiO_2, and (c) sapphire substrates with varying the r.f. power of 150-250W.

In order to compare the structural properties of ZnO films on Si, SiO_2, and sapphire substrates, we have deposited ZnO films with varying the r.f. power for each substrate. Fig. 2 shows the relative intensity of ZnO film with the (002) orientation, deposited on Si, SiO_2, and sapphire substrates, as a function of r.f. power ranging from 150 to 250 W, indicating that the relative intensity of ZnO(002) diffraction peak is significantly higher when ZnO films are deposited on sapphire substrates (Altough the ZnO(002) peaks in case of 250W are not clearly noticeable in Fig. 2, our raw data confirm the above observation). Since the (002) diffraction peaks were distinguishably observed in the grown ZnO films, we surmise that the c-axis oriented ZnO film is obtained, regardless of the substrate material.

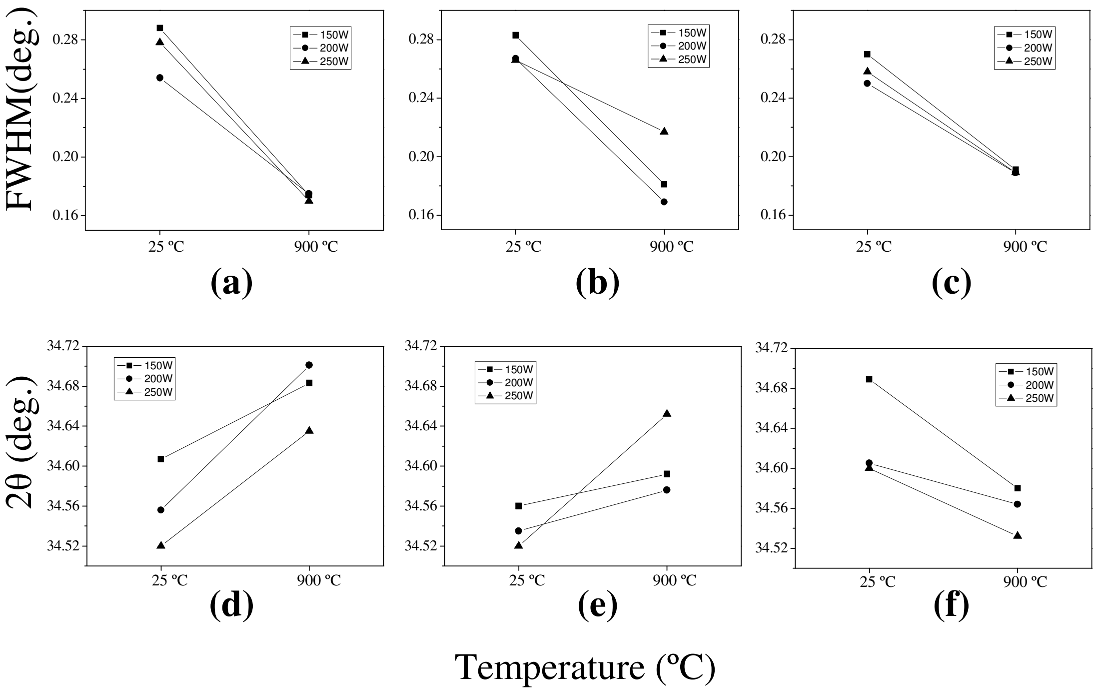

Fig. 3. (a,b,c) FWHM and (d,e,f) peak angle (2θ) of (002) diffraction peak from ZnO film deposited on (a,d) Si(100), (b,e) SiO_2, and (c,f) sapphire substrates with an r.f. power of 150-250 W.

In order to scrutinize the effect of substrate on the structural properties of ZnO films, we have annealed the samples at a temperature of 900°C. Fig. 3 show the full width at half maximum (FWHM) and the peak angle (2θ) of (002) diffraction peak measured in ZnO thin films when deposited with the r.f. power in the range of 150-250W. Fig. 3a-3c indicates that the FWHM decreases by the thermal annealing, regardless of the r.f. power and the substrate material. Since the grain size is inversely proportional to the FWHM, this observation indicates that the grain size becomes larger and thus the crystallinity may be improved by the thermal annealing.

Fig. 3d-3f shows the the 2θ angle of the ZnO films with respect to the (002) diffraction peak. The 2θ angles of the ZnO films deposited in this work are above 34.5°, regardless of the r.f. bias and the substrate material. Since the XRD angle (2θ) of the bulk ZnO are 34.44° and by calculating with the Bragg law [11], proving that the 2θ angle is inversely proportional to the lattice constant, we reveal that c-axis lattice constant becomes larger when deposited at room temperature, regardless of the r.f bias and the substrate material. The XRD data indicate that the 2θ angle increases by the thermal annealing for the ZnO films grown on Si or SiO_2 substrates but decreases by the thermal annealing for the films grown on sapphire substrates. The different substrates will lead to different crystal mismatch and thermal expansion between substrates and ZnO films, and the crystal lattice mismatch and difference of thermal expansion may cause different strains and stress in the films. In case of ZnO films on Si or SiO_2 substrates, we surmise that increasing the annealing temperature increases the compressive stress along the c-axis direction and thus reduces lattice plane spacing and increases the 2θ angle of XRD. On the other hand, in case of ZnO films on sapphire substrates, increasing the annealing temperature reduces the compressive stress along the c-axis direction and thus increases the lattice plane spacing and reduces the 2θ angle of XRD. Further systematic study is necessary to reveal the detalied mechanism.

Summary

ZnO thin films have been grown on Si, SiO_2, and sapphire substrates at room temperature using the r.f. magnetron sputtering technique. The SEM images revealed that the grain size is larger for ZnO films grown on sapphire substrate. The XRD of ZnO films showed c-axis orientation, regardless of substrate material. The relative intensity of (002) diffraction peak is higher for ZnO films grown on sapphire substrates than for ZnO films on Si or SiO_2 substrates. The FWHM of (002) diffraction peak decreases by the thermal annealing, regardless of substrate material. The XRD (002) peak angle of ZnO films on Si or SiO_2 substrates increases by the thermal annealing, while the peak angles in case of sapphire substrates decreases by the thermal annealing. We believe that the approach to investigate the very low temperature growth and annealing behavior of ZnO films depending on the substrate material are a step toward the efficient production of ZnO devices.

Acknowledgements

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