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Structural characterization of ZnO films grown on SiO₂ by the RF magnetron sputtering

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Abstract

We have deposited the ZnO thin films on SiO₂ substrate using RF magnetron sputtering method and have investigated the effect of annealing on the structural quality of ZnO films in the range of 600–1050°C. Annealing at higher temperature was found to enhance the *c*-axis orientation and the grain size of ZnO films.

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In recent years, ZnO thin films have been extensively studied for surface acoustic wave (SAW) devices, optical wave-guides, and transparent conducting coatings [1]. Since the ZnO has a wide band gap of 3.37 eV, low power threshold for optical pumping at room temperature and UV emission resulting from a large exciton binding energy of 60 meV, can be used as light emitting diodes (LED), photodetectors, electroluminescence devices and the next generation UV laser.

ZnO films have been grown by various deposition methods, such as sputtering [2], sol-gel process [3], spray pyrolysis [4,5], pulsed laser deposition [6,7], ion beam deposition [8], plasma enhanced chemical vapor deposition (PECVD) [9], atomic layer deposition (ALD) [10], filtered cathodic vacuum arc technique [11], evaporation

[12], metal-organic chemical vapor deposition (MOCVD) [13,14], and molecular beam epitaxy (MBE) [15].

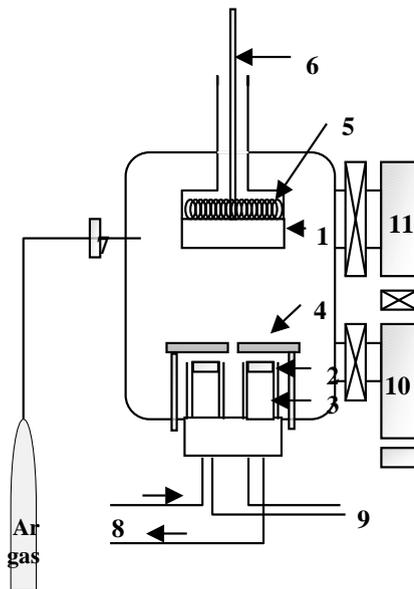
High-quality ZnO films grown on Si substrate pave the way for integration of devices with Si IC technology and amorphous substrates such as SiO₂ and glass substrate have obvious technological advantages and potential applications [16]. Also, the magnetron sputtering is a preferred technique due to its potential for low-temperature processing [17]. Although many researchers have grown ZnO films on sapphire substrates, there are not many reports on growing ZnO thin films on SiO₂ substrate by RF magnetron sputtering method. In this study, we deposit the ZnO thin film on SiO₂ substrates using the RF magnetron sputterer and investigate the effect of annealing temperature on the structural quality of ZnO thin films.

The SiO₂ layer was thermally grown on the Si(100) substrate with a thickness of 60 nm.

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Before loading into the reactor, the substrate was cleaned in acetone for 10 min then rinsed by deionized water for 1 min. In this experiment, we have used a ZnO (99.99% purity) target with a diameter and a thickness of 75 mm and 60 mm, respectively. Fig. 1 shows a schematic diagram of the RF sputtering system used in our experiments. The sputtering was carried out in an Ar (99.99% purity) gas atmosphere by supplying 300 W RF power at a frequency of 13.56 MHz. The flow rate of the Ar gas was set to 30 sccm. The distance between target and substrate was about 80 mm. The ZnO film was grown at 200°C temperature at a pressure of 5.0×10^{-2} Torr and deposition was carried out for 30 min. After RF magnetron sputtering, the samples were annealed in a furnace using quartz tube reactor at temperatures 600°C, 750°C, 900°C, and 1050°C in an oxygen or an air ambient. The structural characteristics of the films were analyzed by X-ray diffraction (XRD) using CuK α 1 radiation ($\lambda = 0.154056$ nm) and by scanning electron microscopy (SEM) (Hitachi S-4200).



1. Substrate 2. Target 3. Magnetron 4. Shutter 5. Heater
6. Thermocouple 7. MFC 8. Cooling system 9. DC power supply
10. Rotary pump 11. Turbo pump

Fig. 1. Schematic diagram of RF magnetron sputtering system.

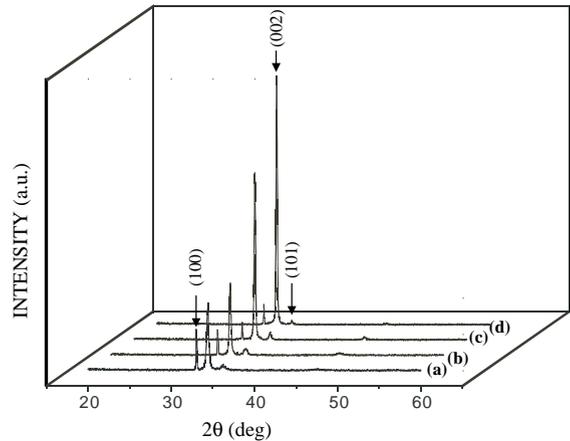


Fig. 2. XRD patterns of ZnO thin films on SiO₂ substrate (a) without annealing and with annealing in an oxygen ambient at (b) 600°C, (c) 750°C, and (d) 900°C.

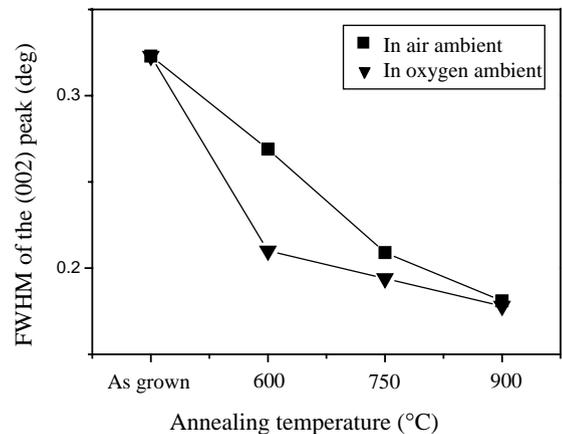


Fig. 3. Variation of the full-width at half-maximum (FWHM) of the ZnO (002) diffraction peak by varying the annealing temperature.

Fig. 2 shows the θ - 2θ XRD patterns of the ZnO thin film on SiO₂ substrate annealed in oxygen ambient for different annealing temperatures. Since the relative intensity of the ZnO(002) diffraction peak, compared to the neighboring (100) and (110) peak, increases gradually by increasing annealing temperature, we observe that the *c*-axis orientation of ZnO films increases by increasing annealing temperature in the range of 600–900°C. The annealing effect in air ambient shows a similar behavior in Fig. 2 (not shown here).

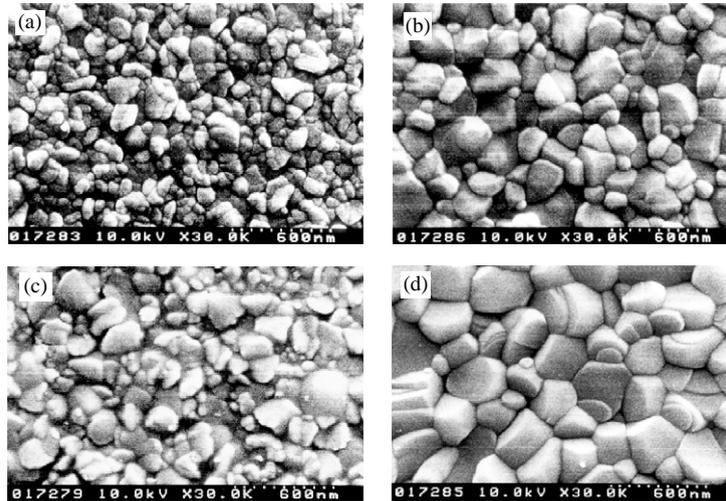


Fig. 4. Plain-view SEM images of ZnO film grown on SiO₂ substrate at the annealing temperature of: (a) 600°C, (b) 750°C, (c) 900°C, and (d) 1050°C in an oxygen ambient.

Fig. 3 shows the FWHM of the (002) diffraction peak according to the annealing temperature in an ambient of oxygen and air, respectively. The FWHM decreases by increasing the annealing temperature. Since the FWHM of the (002) diffraction peak is inversely proportional to the grain size of the film, we see that the grain size of the ZnO thin film increases by increasing the annealing temperature. Investigation of the XRD patterns of the ZnO film indicates that FWHM of the (002) diffraction peak becomes less than 0.3° by annealing at a temperature of 600–900°C.

Fig. 4 shows the plan-view SEM image of ZnO thin films, showing that the grain size becomes larger by increasing annealing temperature and this result agrees with the XRD analysis. We surmise that at high temperature, the atoms have enough diffusion activation energy to occupy the correct site in the crystal lattice and grains with the lower surface energy will become larger at high temperature. Then the growth orientation develops into one crystallographic direction of the low surface energy, leading to the improvement of ZnO crystallinity. We also indicate in this study that annealing effects are not dependent on the ambient gas and further systematic study is underway to investigate the gas effect.

In summary, we have deposited the ZnO thin film on the SiO₂ substrate using the RF magnetron

sputtering technique and have investigated annealing effects in the range of 600–1050°C. The XRD data reveal that the relative intensity of ZnO(002) peak increases by increasing the annealing temperature. The SEM images indicate that the grain size of ZnO film increases by increasing the annealing temperature. The FWHM of the ZnO(002) diffraction peak is less than 0.3° after the annealing treatment.

Acknowledgements

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