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Scaling behavior and coarsening transition of annealed ZnO films on Si substrate

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ABSTRACT

Using reactive radio frequency magnetron sputtering, ZnO films were deposited on Si (001) substrate at room temperature and were annealed at different temperatures ranging from 300 to 1000 °C in air. The annealing behavior has been studied by analyzing morphological and structural evolution of ZnO films quantitatively. A coarsening transition is found occurring at a temperature of about 790 °C. For the annealed films above and below the temperature, the diffusion mechanisms of oxygen vacancies and zinc interstitials are assigned to be responsible for the coarsening behaviors, respectively.

1. Introduction

Since the reports of room-temperature lasing [1–3] and p-type conduction doped with transition metals [4–6], ZnO has received much attention due to its potential applications in optoelectronic devices in blue and ultraviolet spectra of light. Many efforts have been devoted to improve the crystalline quality and optical properties of ZnO films. As one of effective methods, thermal annealing has been widely adopted to enhance the photoluminescence (PL) of ZnO films. It was found that crystallinity of the annealed ZnO films was improved and columnar structures with *c* axis were preferentially formed due to thermal annealing [7]. A Zn–Si–O amorphous layer was observed at the interface between ZnO and SiO_2 for the system of ZnO/Si annealed at 900 °C [8]. Shan et al. reported that the deep-level emission could be increased when the films were annealed in N_2, but decreased when the films were annealed in O_2 [9]. The deep-level emission was assigned to be from oxygen vacancies (V_O) instead of zinc interstitials (Zn_i). However, the atomistic mechanism that improves the crystallinity of ZnO films and results in the creation of V_O is still in debate.

In this article, atomic force microscopy (AFM) and transmission electron microscopy (TEM) are applied to study the morphological evolution and grain coarsening of ZnO films deposited at room temperature and annealed at different temperatures ranging from 300 to 1000 °C in air. A technique of scaling analysis is used to characterize the variation of the surface morphology with annealing temperature quantitatively. A kinetic coarsening transition is observed at a temperature between 700 and 800 °C. The activation energies of diffusion corresponding to the two different coarsening stages are determined by fitting the Arrhenius plots related to the grain size. With the activation energies determined, the atomistic mechanisms driving the grain boundaries motion are discussed in the paper.

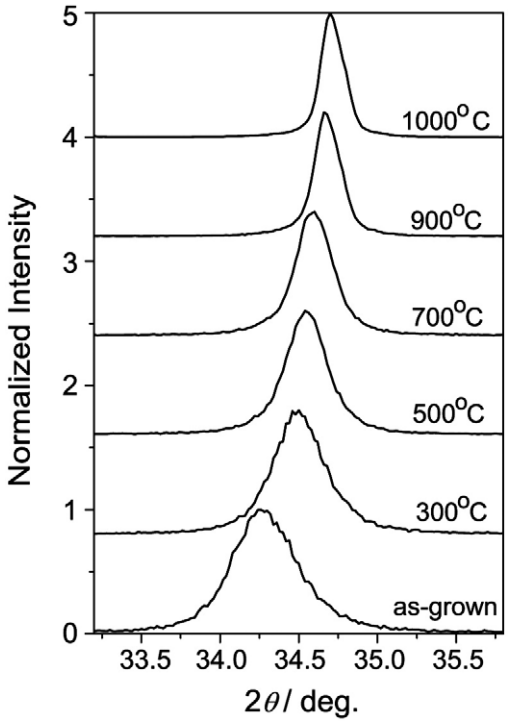

Fig. 1. (002) diffraction patterns of ZnO films at the given temperatures.

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2. Experimental details

ZnO films were grown on Si (001) wafers by reactive rf magnetron sputtering. The Si substrates were cleaned in an ultrasonator with a sequence of acetone, ethanol and deionized water. The sputtering source was a Zn target (99.99%) and the system was pumped to 4.0×10^{-4} Pa before sputtering. The deposition was carried out at 0.5 Pa in an ambient mixture of Ar and O_2 at the flow rate with 20 sccm and 5 sccm, respectively. The power applied to the target was 80 W. Before the deposition, the Zn target was pre-sputtered for 10 min to remove any contamination on the surface. The samples were deposited for 30 min at RT, and annealed at 300, 500, 700, 900, and 1000 °C in air, respectively.

The topological morphology of film surfaces was measured with an AFM operated in contact mode. XRD and TEM were used to examine the crystallinity of ZnO films and the interface features in cross section. PL measurement was carried out at room temperature by using a He–Cd laser at an excitation wavelength of 325 nm. The excitation power was about 30 mW. The PL light was spectrally analyzed by a grating monochromator with a photo-multiplier tube and a lock-in amplifier.

3. Results and discussions

XRD analysis shows the as-grown and annealed films all have (002) and (004) diffraction peaks only, indicating the films are highly (001) textured. The (002) diffraction of the as-grown film peaks at 34.28°, corresponding to the lattice constant along *c*-axis direction $c=0.5232$ nm. The value is much larger than the lattice constant of

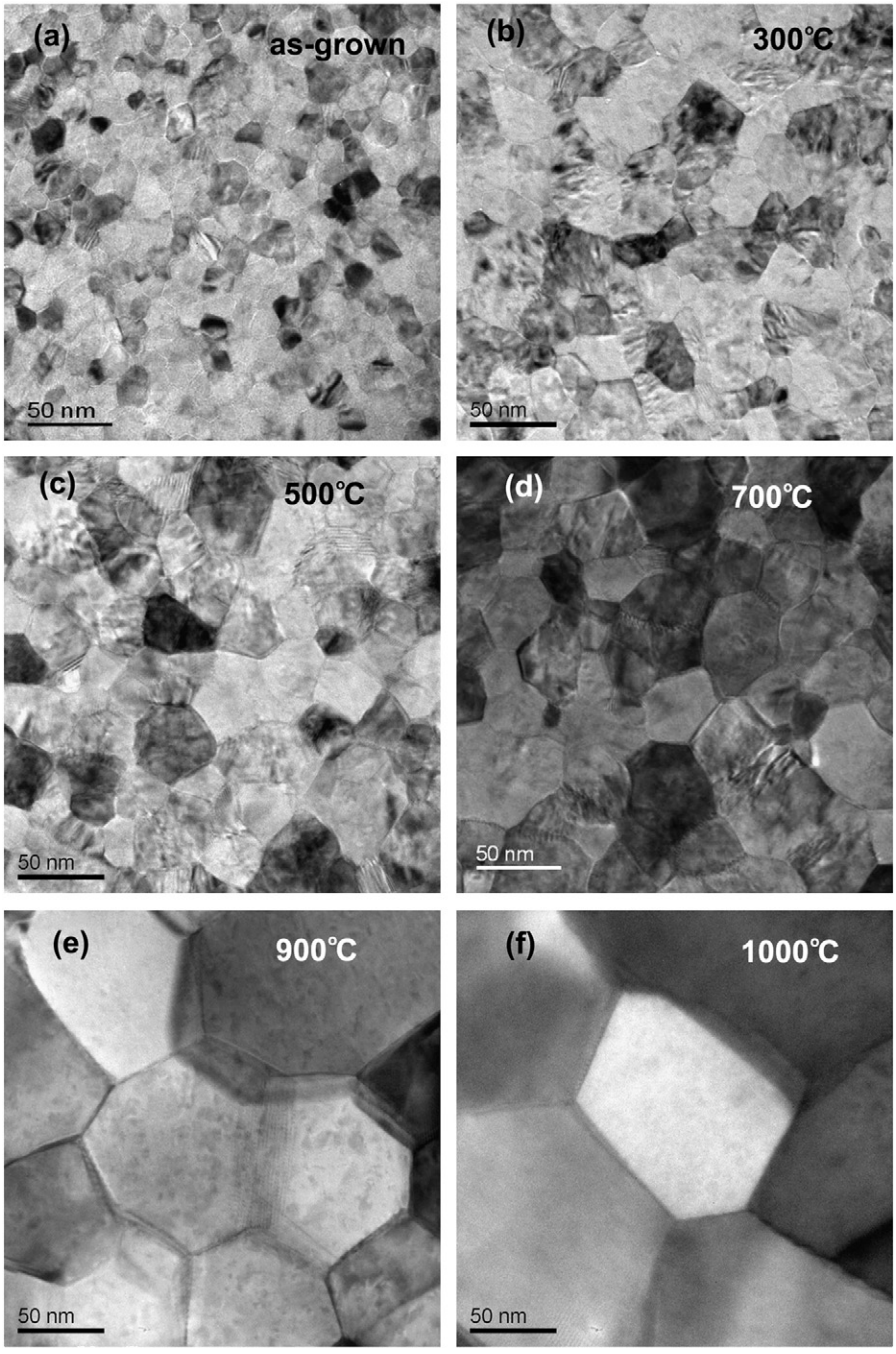

Fig. 2. Plan-view TEM images of the films at the given temperatures.