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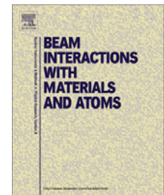

Nanoscratch study of ZnO thin films deposited using radio frequency magnetron sputtering

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ABSTRACT

In this study, a radio frequency magnetron sputtering system was used to deposit zinc oxide (ZnO) thin films onto langasite substrates. The thickness of the ZnO film increased from 0.3 to 1.2 μm upon increasing the deposition power from 100 to 200 W. The predominant growth orientation was along the *c*-axis (0 0 2); the intensities of the signals in the X-ray diffraction spectrum increased significantly upon increasing the film thickness. Scanning electron microscopy images revealed columnar structures in the ZnO films and the morphology of ZnO grains is found to be continuous and dense. It is attributed that oxygen chemisorbs on the target and cases a surface layer of adsorbed oxygen. We suggest that the more neutral ion bombardment on the growing film which induces the higher sputtering rate of the growing film. From *in situ* imaging of scratched tracks, measurement of the coefficient of friction was an effective means of detecting the occurrence of structural defects in the microstructures. We also found that the chemical compositions of ZnO films prepared under various deposition powers could be investigated using X-ray photoelectron spectroscopy.

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1. Introduction

Zinc oxide (ZnO) has recently emerged as a promising alternative to gallium nitride because of its large band gap and large excitation binding energy [1]. ZnO thin films are mainly used as transparent conductive films in solar cell windows. The successful fabrication of semiconductor devices incorporating ZnO will require a better understanding of its mechanical characteristics and its optical and electrical properties; indeed, contact loading during processing or packaging can significantly degrade the performance of such devices [2–4].

The mechanical properties of ZnO films have been investigated widely [5–8]. In particular, nanoscratch and nanoindentation techniques have become important tools for analyzing ZnO thin films of various compositions [9,10]. The nanoscratch technique can be used to characterize the nanotribological properties of a sample; the process involves scratching its surface with a diamond tip and recording the coefficient of friction. Although the mechanical properties of ZnO films have been discussed widely [5–11], the issue about nanotribology of ZnO films is less well understood; therefore, precise studies are required if ZnO films are to be used as structural/functional elements in devices.

In this study, the growth and characteristics of ZnO films deposited using a radio frequency (RF) magnetron sputtering system were investigated at various deposition powers. The characteristics (crystallinity, morphology, roughness, and nanotribology) of these ZnO films were measured using various techniques, including X-ray diffraction (XRD), field emission scanning electron microscopy (FESEM), atomic force microscopy (AFM), and nanoscratch techniques. The effect of the deposition power on the microstructures and nanoscratch properties was also investigated.

2. Experimental descriptions

An r.f. magnetron sputtering system using a water-cooled 4 inch-diameter Zn target (99.999%) in an Ar/O_2 gas mixture was employed to deposit ZnO films onto langasite (La3Ga5SiO14, LGS) substrates (substrate–target distance: 5 cm); r.f. powers of 100, 150, and 200 W were used for the deposition of three ZnO samples, respectively. In addition, the base pressure was 6×10^{-6} torr, the sputtering pressure was 3–9 mtorr, the substrate temperature was set at 200 °C, the $O_2/(Ar + O_2)$ ratio was set at 20% (purity of O_2 gas: 99.995%; purity of Ar gas: 99.995%), and the duration of the deposition was 1 h. After deposition, cross-sectional images of the ZnO films were recorded using a JEOL JSM-7001F field emission scanning electron microscope, which revealed that the thickness of the ZnO films increased regularly from 0.3 to 1.2 μm upon increasing the deposition power from 100 to 200 W. The deposition rates at 100, 150, and 200 W were, therefore, calculated to be ca. 5.0, 12, and 20 nm/min, respectively.

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To determine the morphological surface properties, the samples were scanned using a Digital Instruments Nanoscope III atomic force microscope operated at a constant scan speed (1 μm/s), with a constant load (30 nN) applied to the cantilever. The nanotribological properties of the samples were measured through nanoscratch tests, using a combination of AFM and a nanoindentation measurement system (Hysitron, Inc.), operated at a constant scan speed (2 μm/s) and constant load (5000 μN) and featuring a diamond Berkovich indenter tip (radius: 50 nm); scratching was performed at a constant scratching speed (0.13 μm/s) over a constant length (10 μm). Surface profiles before and after scratching were obtained by scanning the tip with a sufficiently small load that did not produce a measurable displacement. After scratching, the wear tracks were imaged using AFM. All tests were realized at least five times to determine the reproducibility; the data were averaged using the central regions of the scratches (i.e., neglecting any steps, peaks, or depressions that may have arisen).

3. Results and discussion

The ZnO films were polycrystalline, revealing (1 0 0), (0 0 2), and (1 0 1) peaks for hexagonal ZnO at 31.75, 34.35, and 36.31°, respectively, in their XRD spectra (Fig. 1). After deposition at 100 W, weak diffraction peaks were observed for ZnO, suggesting the onset of ZnO growth. The spectrum obtained after deposition at 150 W revealed the presence of pure and polycrystalline ZnO having a wurtzite structure [12]. The intensity of the (0 0 2) peak increased significantly upon increasing the deposition power to 200 W, revealing enhanced crystallinity in the (0 0 2) oriented plane. The average grain size in a film can be estimated using Scherrer's equation [13]:

$$D = 0.9\lambda/\beta\cos\theta$$

where D is the mean dimension of the crystallites, λ is the X-ray wavelength, and β is the full width at half-maximum (FWHM) of the (0 0 2) peak. The average grain size in the films decreased from 109 to 104 to 98 nm upon increasing the deposition power from 100 to 150 to 200 W, respectively. Lin et al. [14] reported that grain overgrowth caused by a high substrate temperature will induce a rough surface; they also suggested that the surface structure of a ZnO film is influenced by the use of zinc or oxygen support. Similar phenomena have been reported [15,16] for ZnO films deposited through dc magnetron sputtering at various temperatures, resulting in preferred orientation along the (0 0 2) plane. Thus, the average grain size is not controlled by a single parameter.

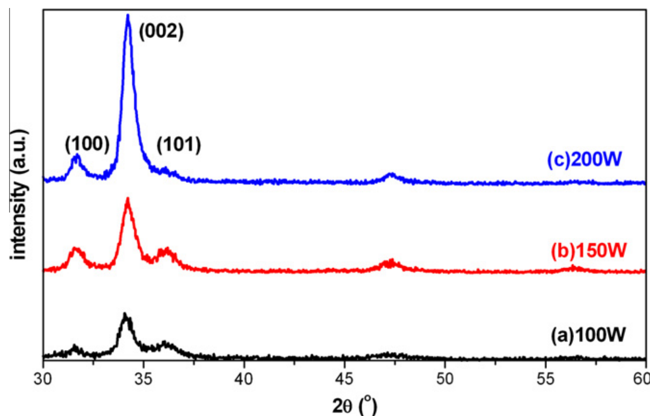

Fig. 1. XRD spectra of ZnO thin films deposited at deposition powers of (a) 100 W (weak diffraction peaks of ZnO), (b) 150 W (presence of pure and polycrystalline ZnO), and (c) 200 W (improvement of crystalline quality with power).

Fig. 2 displays SEM and AFM images representing the surface morphologies and 3D topographies of ZnO films prepared at deposition powers of 100, 150, and 200 W. It is clear that no specific epitaxial orientation occurred on the langasite substrate surface, leading to randomly oriented nuclei. The insets reveal cross-sectional SEM images of the ZnO films, the thickness of which increased regularly from 0.3 to 1.2 μm upon increasing the deposition power from 100 to 200 W. Columnar structures were observed in the ZnO films; their possible mechanism of formation has been reported previously [15,16]. The Zn and O compositions during the sputtering process were also dependent on several parameters [15–17]. Lu et al. reported [18] that the growth rates of ZnO films increase upon increasing the r.f. power and that the resulting ZnO grains were continuous and dense. In our experiments, however, the intensity of the c-axis (0 0 2) peak increased sharply upon increasing the deposition power from 100 to 200 W, while the other two peaks coexisted with much smaller intensities and greater FWHMs. Although an increased peak intensity in an X-ray diffractogram might be due to a larger thickness of the films, it might also be attributable to indirect crystallinity as a result of the higher deposition power. We suspect that under a high deposition power, more Zn atoms can reach the substrate and have more opportunity to form nuclei. In other words, more nuclei sites will be generated and a large number of small grains will grow, leading to a smaller-grain structure and a higher growth rate.

The height roughness parameter (R_a) and the root mean square roughness (R_{ms}), determined from AFM images, can be used to quantify the morphologies of ZnO films [19]. Fig. 2 provides typical images obtained using the various growth procedures; the surface roughnesses and apparent feature sizes are clearly evident. The ZnO film formed at 100 W was smooth (R_{ms} = 11.3 nm); it roughened to values of R_{ms} of 14.5 and 16.5 nm for deposition powers of 150 and 200 W, respectively; i.e., the surface roughness increased rapidly upon increasing the deposition power from 100 to 200 W. Although we obtained appropriate grain sizes and flat surface crystals, the deposition conditions affected not only the grain growth but also the Zn and O compositions during the sputtering process. When the content of supporting O atoms was increased [14], the surface morphologies of the films displayed greater roughness. Table 1 summarizes the values of R_a and R_{ms} obtained under the various growth conditions. For oxygen content in the range of 20%, rapid oxidation of the target takes place at high deposition power; this increase in secondary electron yield causes more ionization of the sputtering gas, therefore increase the sputtering rate. It is attributed that oxygen chemisorbs on the target and cases a surface layer of adsorbed oxygen. We suggest that the increased surface roughnesses based on the more neutral ion bombardment on the growing film which induces the higher sputtering rate of the growing film.

AFM studies of the scratched tracks revealed more details concerning the scratching process. Fig. 3 displays AFM images of scratched tracks formed on the ZnO/LGS samples prepared under the various deposition conditions. Fig. 3a reveals the presence of a significant creep in the ZnO film prepared at 100 W, associated with plastic deformation of the material. Under a constant normal load of 5000 μN, a plowing scratch track appeared without debris on the sides of the scratch, likely to be the cause of the lower coefficient of friction (μ = 0.1 ± 0.01). Notably, the ZnO/LGS films exhibited lower surface roughness for lower values of μ. For the ZnO film sample prepared at 150 W Fig. 3b, in addition to plowing scratch tracks, pile-up appeared on the sides of the scratches (μ = 0.175 ± 0.01). After deposition at 200 W Fig. 3c, lateral sliding emanating from the indenter edge occurred, leading to removal of the film through a ploughing mechanism; scratch debris or nanoparticles appeared inside or around the nanoscratch tracks at a

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