한국표면공학회지
Journal of the Korean Institute of Surface Engineering
Vol. 28, No. 6, Dec. 1995
<연구논문>

Crystallographic Characteristics of ZnO Films Deposited on SiO_2/Si Substrate

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Abstract

The RF planar magnetron sputtering technique was used to fabricate uniform $ZnO/SiO_2/Si$ thin films at high growth rate. A detailed crystallographic character of these thin films has been carried out using XRD, XRC, and SEM. These thin films have the configuration of c-axis orientation perpendicular to SiO_2 / Si substrate. The dependence of the thickness of $ZnO/SiO_2/Si$ films on applied RF power parameters was also investigated. The crystallinity of films was improved as the substrate temperature was high, RF input power increased, and Ar/O_2 ratio decreased. Also, most of ZnO films fabricated on SiO_2/Si were suitable for SAW filter since a standard deviation of XRC (002) peak was less than 6°. The presence of the SiO_2 layer has a beneficial effect on the crystalline quality of the grown ZnO films.

1. Introduction

There has been substantial interest in deposition of highly oriented piezoelectric thin films on insulating and semiconductor substrates for signal processing applications. Zinc oxide has been recognized as one of the most excellent piezoelectric materials due to its high electromechanical coupling factor[1)]. Owing to these characteristics, ZnO is suitable for ultrasonic transducers especially in a high-frequency region. Although it is difficult to obtain large single-crystal of ZnO, piezoelectric thin films of ZnO can be grown on various substrates[2)]. High quality ZnO films have usually been deposited on silicon or glass substrates. This paper is concerned with the influence of the applied RF power, sputtering atmosphere and substrate temperature on the c-axis orientation and other crystallographic characteristics of $ZnO/SiO_2/Si$ films. The quality of $ZnO/SiO_2/Si$ films studied by X-ray diffraction(XRD), scanning electron microscopy (SEM), X-ray rocking cruve(XRC) is reported. Thin SiO_2 buffer layer thickness is 2400Å.

2. Experiment

Fig. 1 shows a schematic diagram of the RF planar magnetron sputtering system used in this study. A 2 inch-diameter disk of hotpressed

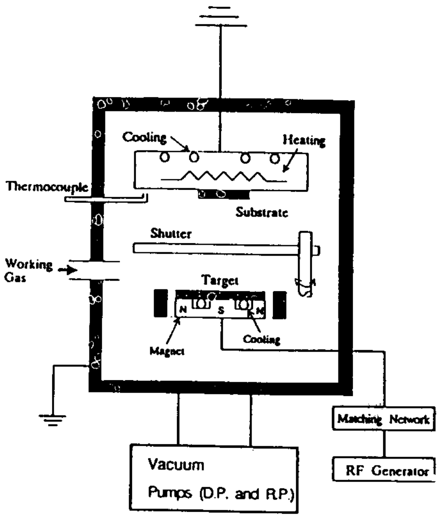

Fig. 1. A schematic diagram of the RF planar magnetron system.

Table 1. Sputtering condition for preparation of ZnO films

Target	ZnO(CERAC-318 U.S.A)
Substrate	SiO_2/Si(100)
Target-Sub distance	5cm
RF power	100W-500W
Ar/O_2 ratio	100/100, 200/100, 300/100, 400/100, 500/100(mℓ/min)
Substrate temperature	150℃-400℃
Sputtering pressure	2×10^{-3} torr
Sputtering time	30min

ZnO powder (99.9% purity, Cerac-318 U.S.A.) was used as a target. The substrate used in this experiment was a Si(100) with a thin SiO_2 buffer layer (2400Å thick). Introduction of a SiO_2 buffer layer between ZnO and Si would promote the adhesion of those films. That is, ZnO/SiO_2/Si thin film shows better thermal stability than the ZnO/Si. Prior to metal clamping on the holder, the substrate was cleaned by a standard cleaning procedure to eliminate an organic matter from the surface. Cleaning was carried out for 5 minutes in the ultrasonic cleaner in the order of T.C.E. Aceton, Methanol, and deionized water, respectively. Before the deposition, the sputtering chamber was pumped down to 2×10^{-6} Torr using diffusion pump in order to remove any impurities. The substrate temperature was controlled by a temperature controller with K-type thermocouple. Table 1 shows the condition of deposition. First, ZnO thin films were deposited at the RF power of 100W, Ar/O_2 ratio of 200 mℓ / 100 mℓ, and the substrate temper ature ranging from 150℃ to 400℃. Second, at the fixed temperature and Ar/O_2 ratio that we obtained a thin film of good crystallinity, sputtering was performed with the increase of RF power. Last, at a fixed RF power 200W and substrate temperature 400℃, sputtering was carried out with the variation of Ar/O_2 ratio. The crystallinity of thin films was analyzed by XRD where CuKα was used and wavelength and the diffraction angle range were 1.5418Å and 30°~40°, respectively. The configuration of c-axis orientation perpendicular to the substrate surface was studied by means of a standard deviation obtained from XRC of ZnO (002) plane and to determine the thickness of ZnO thin films SEM was used.

Ⅲ. Results and discussion

Fig. 2 shows the changes in deposition rate of the sputtered ZnO films as a function of RF power, substrate temperature, and Ar/O_2 ratio. The deposition rate of ZnO thin films increased as the applied RF power increased. In addition, the deposition rate independent of the substrate temperature and the Ar/O_2 ratio. These results indicate that the deposition rate was independent of the reaction rates at the substrate surface but dependent on the number of Zn and O atoms reaching the substrate. Therefore, the higher RF input power results into an increased sputtered particles[3]. Figure 3 shows X-ray diffraction patterns of ZnO thin films deposited on SiO_2/Si substrates under various conditions shown in Fig. 3(a), (b), and (c). It should be mentioned that non c-axis orientation peaks such as the (100) and (101) plane peaks appear at high RF power (300W, 500W), Ar/O_2 rate (300/100, 400/100, 500/100), and the substrate temperature of 150℃. SEM was employed to investigate the microstructure and thickness of the deposited films.

Fig. 4 shows a cross-sectional view of the ZnO thin film deposited at RF power of 500W, Ar/O_2 ratio of 200mℓ/100mℓ, and the substrate temperature of 400 ℃. Its cross section also

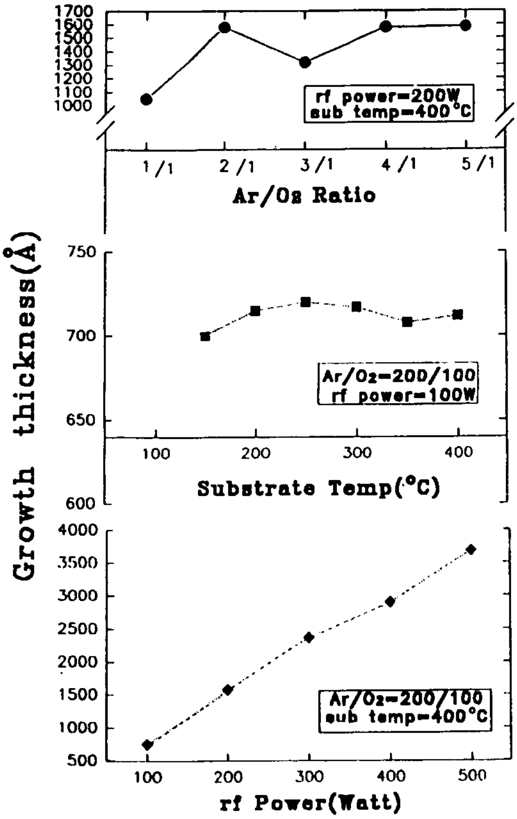

Fig. 2. Relationship between ZnO thickness and RF power Substhate Temperature, Ar/O_2 ratio for 30min deposition.

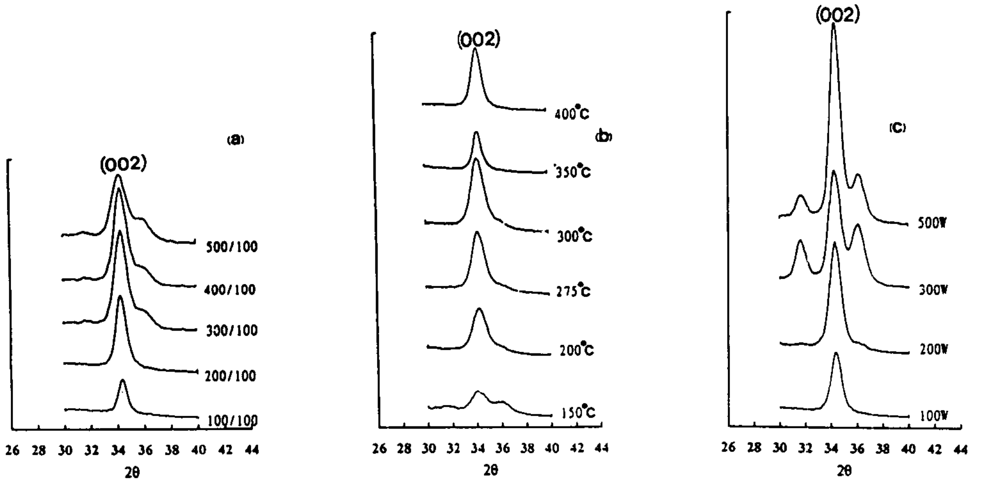

Fig. 3. X-ray diffraction pattern of the (002) peak of a c-axis oriented ZnO film. The sample (a) were grown at a substrate temperature 400℃ with 200W RF power, sample (b) were grown at a gas ratio of Ar : O_2=2 : 1 with 100W RF power and sample (c) were grown at a substrate temperature of 400℃ with a gas ratio of Ar : O_2=2 : 1

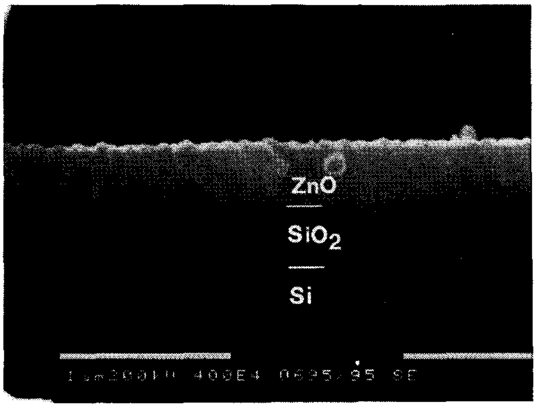

Fig. 4. SEM cross-section of the $ZnO/SiO_2/Si$ specimen.

shows the film on SiO_2 buffer layer consisting of the columnar structure. Obviously, the film exhibits the characteristics of fine and dense grain structures with a smooth interface between the Zno film and SiO_2/Si substrate. In order to examine the distribution of the c-axis orientation (002), X-ray rocking curves of the film was measured in the following way : first, set a substrate face with a ZnO film in paralled to the reference plane of a diffractometer. Adjust separately the θ and 2θ axis of the diffractometer to the angles where the X-ray detector gives the maximum output for the (002) peak. The angles should be near 17.70° and 34.4°, respectively, Then fix the 2° axis, that is, the detector arm at the angle and scan the θ axis, that is the specimen arm around 17.70°. Approximate this as a Gaussian curve to a good approximation[4].

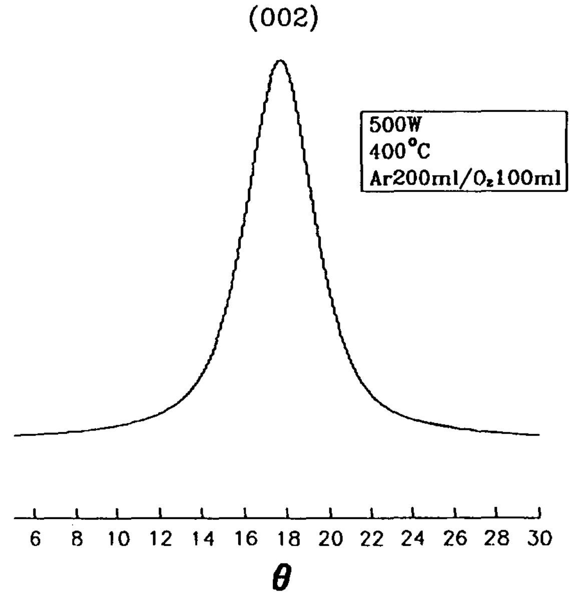

Fig. 5. X-ray rocking curve of the (002) peak.

Fig. 5 shows the rocking curve of the film prepared by a 500W RF input power. The minimun value of standard deviation is 1.64°. The physical structure of the deposited $ZnO/SiO_2/Si$ films was investigated by the full width at half maximun (FWHM), the grain size and c-lattice constant of c-axis (002) orientation from the X-ray diffraction. On the other hand, standard deviation was analyzed by X-ray rocking curve.

Fig. 6 shows the standard deviation, grain size, FWHM, and c-lattice constant as a function of substrate temperature. From Fig. 6 it can be seen that the FWHM, grain size and standard deviation depend on the substrate temperature. At low substrate temperature the surface adatom mobility at the substrate do not have enough energy to establish a well-oriented structure and consequently the FWHM, standard deviation is high value. However, the FWHM, and standard deviation decrease as the substrate temperature increases. As shown in Fig. 6 we can see that ZnO films grown at 350 ℃ have the best quality. The c-lattice constant is almost constant and is independent of the substrate temperature. We obtain the lattice constant along the c-axis of the ZnO films as 5.22Å, whereas that of the ZnO single crystal is

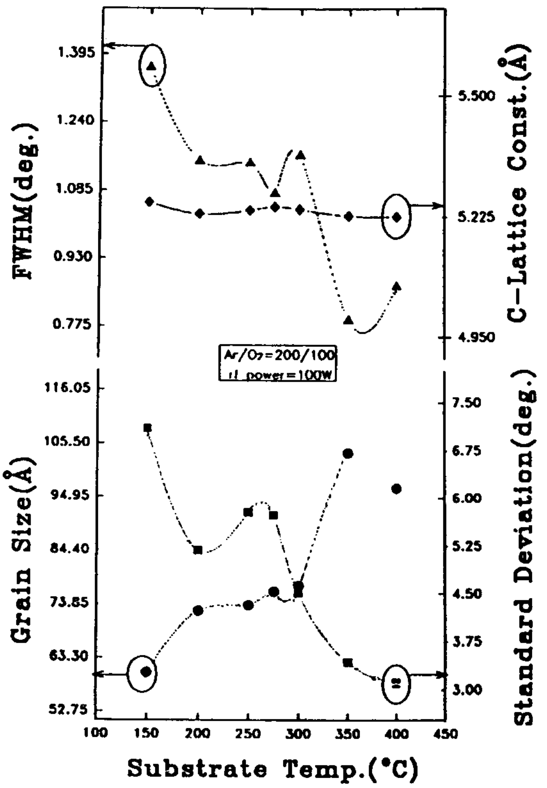

Fig. 6. Crystallographic indices as a function of substrate temperature.

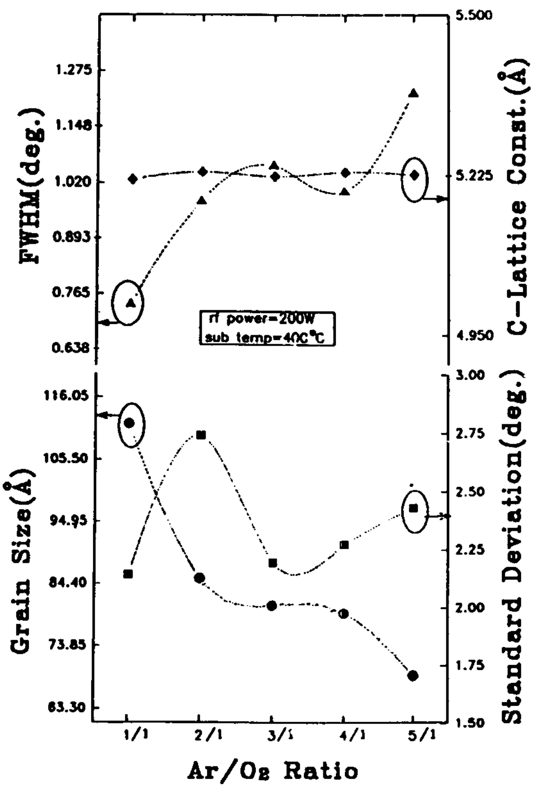

Fig. 7. Crystallographic indices as a function of Ar/O_2 ratio.

5.19Å[1]. Therefore these ZnO films must have a slight lattice strain.

Fig. 7 shows the relationship between the Ar/O_2 ratio and crystalline structure for samples prepared at 400℃, with 200W RF input power. From the experimental results, the standard deviations are not very dependent on the Ar/O_2 ratio. However, associated grain sizes are influenced significantly. In general, when a metal oxide is sputtered in argon for deposition, it is difficult to obtain a complete molecular composition onto any substrate unless a compensatory process is employed. Therefore, to obtain a stoichiometric ZnO thin films, the content of oxygen in a mixed gases of oxygen and argon is a very important factor[5]. When Ar/O_2 ratio was 1 : 1, the value of FWHM was minimum. This indicates that the more Ar atoms exist than O_2 atoms, the more Zn atom sputtered are, so the time necessary for ZnO thin film to crystallize on the substrate surface is fairly insufficient. From the point of view of fine structure, the high quality of ZnO film is not obtained at a large Ar/O_2 ratio. On the other hand, c-lattice constant of ZnO films were not almost changed. Fig. 8 shows the influences of RF power on the FWHM, the standard deviation and grain size. From Fig. 8, it can be seen that the standard deviation depends linearly decrease on the RF power. For highly c-axis oriented ZnO films,

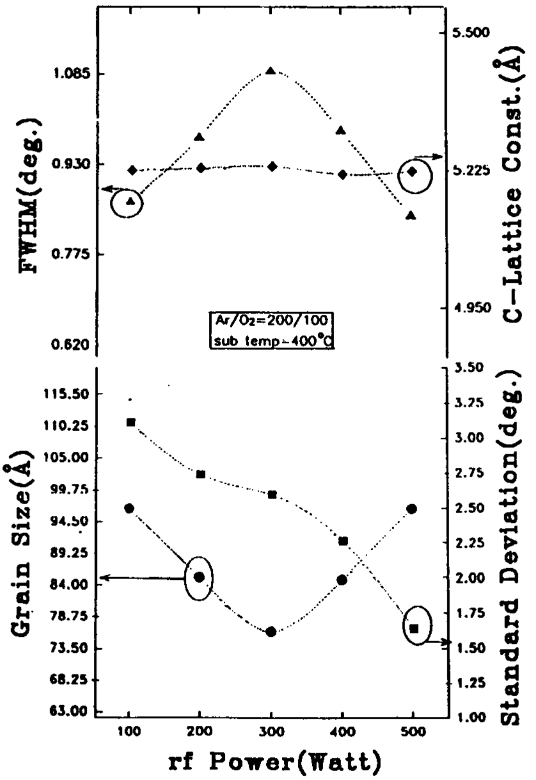

Fig. 8. Crystallographic indices as a function of RF power.

the full width at half maximum intensity (FWHM) of the ZnO (002) peak is smaller, the average grain size obtained from the Scherrer's formula[6] is larger and the standard deviation is smaller than those of the poor-quality films. From the experimental results, the best superior quality of the grown ZnO film is obtained at a RF power of 500W. In this experiment, the c-lattice constant is not dependent on the RF power.

IV. Conclusion

In this paper, the influence of the deposition parameter, such as RF power, substrate temperature, and Ar/O_2 ratio on the crystallinity of ZnO thin films prepared by RF planar-magnetron sputtering has been investigated. From the experimental results, the crystallinity of ZnO thin films was improved as the substrate temperature was high and RF input increased. These resulted in because the reaction of Zn and O atoms on the substrate was promoted by the thermal energy of substrate heating. We could find the optimum conditions for growing ZnO films as follows : the deposition gas composition of Ar/O_2 is 2 : 1, the RF input power is 500W, the substrate temperature keeps at 350 ℃. The thickness of films became larger along with the increase in RF input power but was independent of Ar/O_2 ratio and substrate temperature. The growth rate was independent on the reaction rate at the substrate surface, but dependent on the number of Zn and O atoms reaching the substrate. The minimum standard deviation was 1.6° at substrate temperature of 400℃, RF power of 500W, and Ar(200cc/min) $+O_2$(100cc/min) sputtering gas mixture. The standard deviation depends both on the applied RF power and on the substrate temperature, and the grain size depends both on the applied RF power and on the sputtering gas Ar/O_2 ratio. The c-lattice constant is almost independent of the deposition parameters.

Acknowledgements

This rearch was partially supported by Korea Telecom Research Laboratories.

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