EFFECT OF TARGET TO SUBSTRATE DISTANCE ON THE PROPERTIES OF RF MAGNETRON SPUTTERED ZnO THIN FILMS

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ABSTRACT

We have grown ZnO thin films on silicon substrate by r.f magnetron sputtering using metallic zinc target and investigated the target to substrate distance dependence of their stress and structural properties using X-ray diffraction (XRD). We have confirmed that high-quality ZnO films can be grown in the target to substrate distance (d) range from distance 3 to 9 cm. As the target to substrate distance (d) varied, the microstructure of ZnO films was also varied by the stress with a litle change in crystal orientation. It was found that the compressive stress of ZnO films originated from the small thermalisation of the films.

Keywords: *interactions ZnO; R.F sputtering magnetron; X-ray diffraction; piezoelectric.*

1. INTRODUCTION

The upcoming needs for higher carrier frequencies in MEMS devices open opportunities for bulk acoustic wave (BAW) technology based on piezoelectric thin films. One promising piezoelectric material, which has attracted substantial attention in the recent years, is ZnO. For these needs, several techniques were investigated in order to produce the high quality epitaxial and textured films for these kinds of acoustic transducers. Epitaxial ZnO films have been successfully obtained by metal-organic chemical vapour deposition [1], spray pyrolysis [2], sol-gel method [3], pulsed laser deposition [4], chemical bath deposition technique [5], solution combustion method [6] and sputtering [7-13], and have been employed to obtain ZnO thin films onto different substrates.

The Full Width at Half Maximum (FWHM) of the (002) X-ray rocking curve is known to be suppressed below about 0.5° for obtaining effective electromechanical coupling [8]. The thermal stresses were determined by using a bending-beam Thorton method [13] while thermally cycling films. According to the literature, the reactive sputtering technique has received a great interest because of it's advantages for film growth, such as easy control for the preferred crystalline orientation, epitaxial growth at relatively low temperature, good interfacial adhesion to the substrate and the high packing density of the grown film. These films may exhibit the c-axis preferred orientation and the piezoelectric properties. In FBAR devices, the ZnO film should exert a minimum stress on the underlying layer and also have a high piezoelectric constant. The piezoelectric constant of monocrystalline ZnO films has been determined by the degree of c-axis texturing. However, the very high residual stress often observed in the monocrystalline ZnO thin films appeared as a potential obstacle to investigation.

Sputtered ZnO films often exhibited a large compressive stress that can be reduced by thermal annealing. This can change the film morphology [12], leading the recrystallisation of the films.

In this work, we attempted to make a systematic study of the residual stress and microstructure of reactively sputtered ZnO films as function of the target to substrate distance before and after annealing.

2. MATERIALS AND METHODS

Zinc oxide films were deposited by r.f magnetron sputtering using a zinc target (99,99%) with diameter of 51 mm and 6 mm thick. Substrate is p-type silicon with (100) orientation. The substrates were thoroughly cleaned with organic.

Magnetron sputtering was carried out in oxygen and argon mixed gas atmosphere by supplying r.f power at a frequency of 13.56 MHz. The RF power was about 50 W. The flow rates of both the argon and oxygen were controlled by using flow meter (ASM, AF 2600). The sputtering pressure was maintained at $3.35.10^{-3}$ torr controlling by a Pirani gauge. Before deposition, the pressure of the sputtering system was under 4.10^{-6} torr for more than 12 h and were controlled by using an ion gauge controller (IGC – 16 F).

Thin films were deposited on silicon, substrate under conditions listed in table-1. These deposition conditions were fixed in order to obtain the well-orientation zinc oxide films.

Table 1. Parameters of sputtering deposition of ZnO.

Sputtering pressure	3.35×10^{-3} Torr
Mixture gas	$Ar + O_2$ = 80 – 20 %
Power RF	50 W
Sputtering time	6 h
Substrate temperature	100 °C
Target-substrate distance	3 - 9 cm

The presputtering occurred for 30 min to clean the target surface. Deposition rates covered the range from 0.35 to 0.53 μm/h. All films were annealed in helium ambient at 650°C for 15 mn.

In order to investigate the crystallographic properties of the ZnO films, we carried out an X-ray diffraction (XRD) analysis using CuK_α (λ = 0,154054 nm) radiation. The root-mean-square (rms) surface roughnesses of the films were characterized by using an atomic force microscope (AFM).

3. RESULTS AND DISCUSSION

Figure-1 shown X-ray diffraction (XRD) pattern for samples deposited at several target to substrate distance (ranging from 3 to 9 cm) before and after annealing.

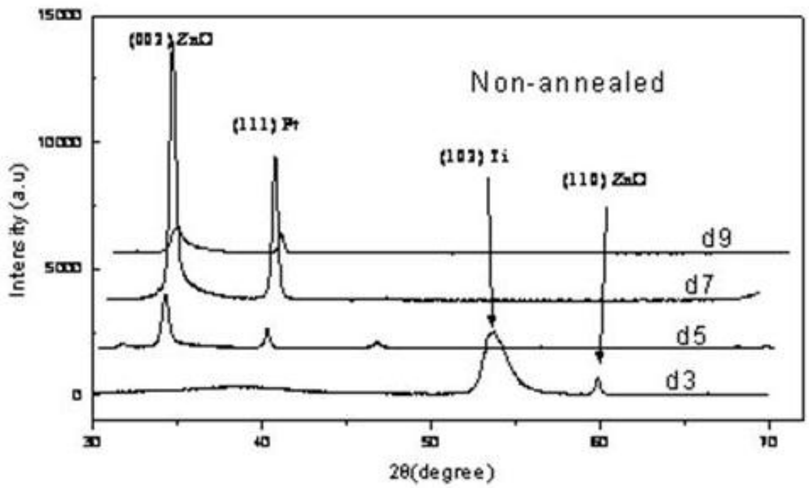

Figure 1a. XRD diagram from non annealed ZnO samples deposited on silicon substrates at different target to substrate distance.

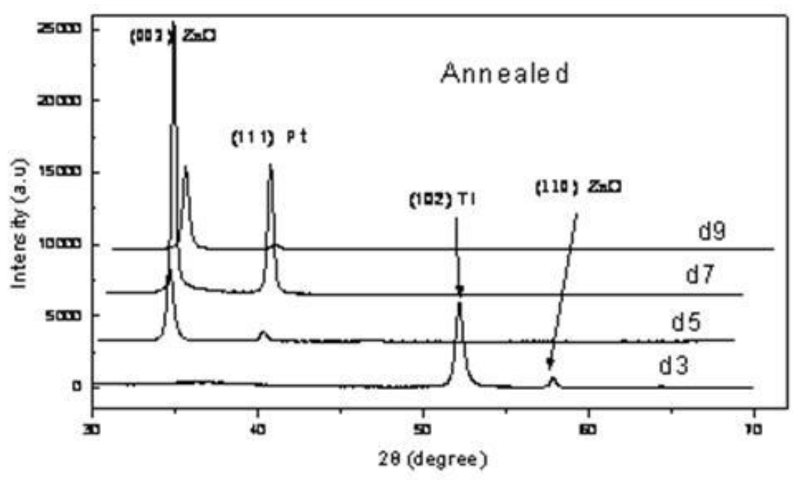

Figure 1b. XRD diagram from annealed ZnO samples deposited on silicon substrates at different target to substrate distance.

The thickness of the films is approximately 3 μm. We believe that the preferred orientation of the film changing from (002) to (110) direction when increased the target to substrate distance. At target to substrate distance below 5 cm, no evidence of (002) peak of ZnO was observed. In target to substrate distance range between 5 and 9 cm, peaks corresponding to the hexagonal structure of ZnO detected. No other peak could exhibit preferred c-axis orientation. The intensity of the (002) peak increased as the target to substrate distance is increased to 7 cm due to the improvement of the films crystallinity. However, at 9 cm, the (002) peak intensity is decreased which indicates that the degree of crystallinity of the films is deteriorated. The diffraction peak of (002) annealed ZnO film was stronger and shaper than that of ZnO film non-annealed. This indicated that the quality of ZnO film was improved after thermal annealing. From the XRD patterns, it is confirmed that the ZnO film quality is gradually improved as the annealing at 650°C temperature.

The figure-2 shown the FWHM of ZnO thin film annealed and non-annealed. The FWHM of ZnO annealed decreased as the target to substrate distance is increased to 7 cm. However, at 9 cm, the FWHM is increased.

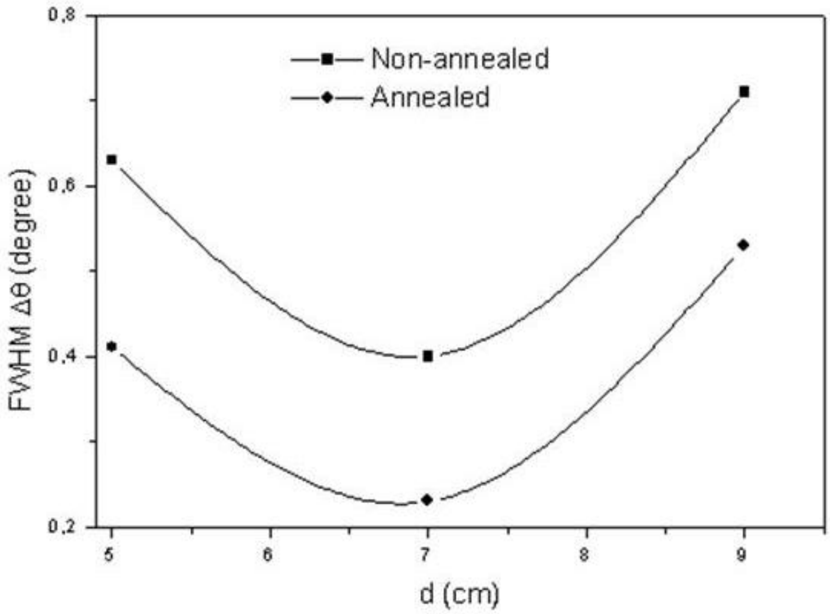

Figure 2.FWHM of (002) X-ray rocking curves for non annealed and annealed ZnO samples deposited on silicon substrates at different target to substrate distance.

These results indicate that the crystal quality of the film deposited decrease for target to substrate distance superior to 7 cm. The above results reveal that a typical FWHM of 0.25° is early obtained when thin films are annealed. This value is substantially lower than the upper limit of 0.5° compared with the minimum reported FWHM of thin films deposited on silicon [14].

Figure-3 shows the variation of stress in the films of ZnO annealed and non-annealed as a function of the target to substrate distance. The compressive residual stresses were evaluated from the XRD spectrum.

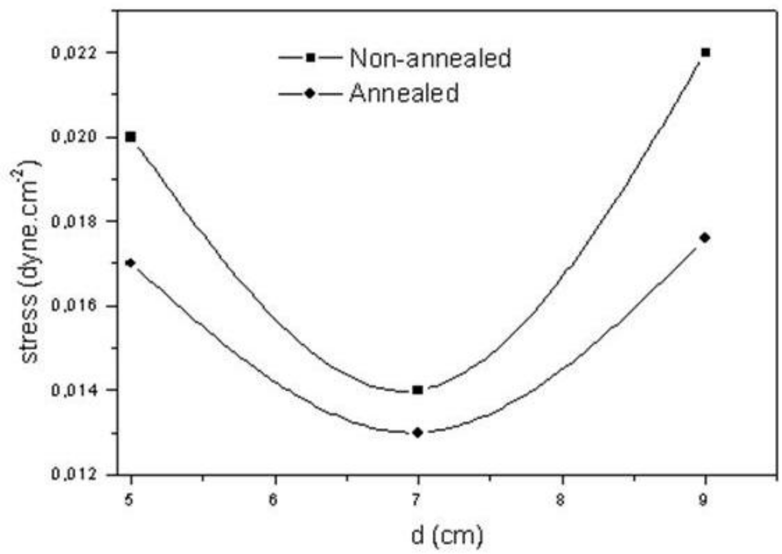

Figure 3. The variation of stress with respect to target to substrate distance of non annealed and annealed ZnO films.

In the ZnO film with a hexagonal structure, the residual film stress in the direction parallel to the surface proportionally acts as a c-axis strain. Although a ZnO film with the preferred orientation is grown at 7 cm, the stress of the films increases with increasing target substrate distance. This stress is not a thermal stress originating from the difference between the thermal expansion coefficients of the film and the substrate, but an intrinsic stress originating from the film structure and density because the film grown at 100 °C. At the temperature of annealing, the relatively little particle scattering within the plasma allows high energy particles to strike the film at an angle approximately normal to its surface. This cause film densification by the voided regions in the microstructure through efficient atomic rearrangement [13]. In the same temperature, the stress is minimal for a target to substrate distance of 7 cm. This result is also caused by the enhancement of nucleation sites with increasing target to substrate distance.

Figure-4 shows the root-mean-square roughness of the ZnO thin films annealed and non-annealed in terms of different target to substrate distance. According to the figure, the minimum roughness of the films is dependent on the crystal structure.

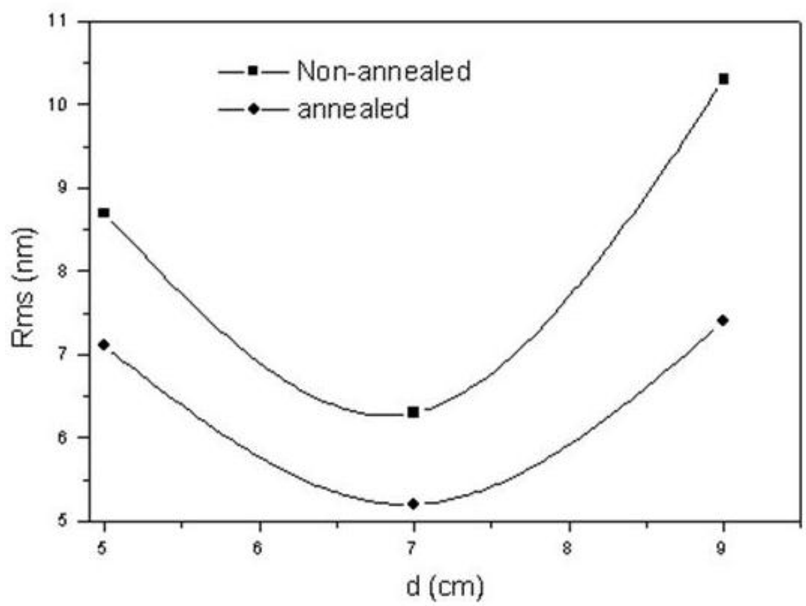

Figure 4. rms of ZnO films annealed and non annealed at different target to substrate distance.

The films deposited at low target to substrate distance are amorphous. We believe that the preferred orientation of the film changing from (002) to (110) direction is responsable for the improved surface roughness. The target to substrate distance used for obtained the high quality of ZnO film is 7 cm. For this distance, the sputtered particles will be completely thermalized and reach the substrate by difusion. Further, the ion bombardment induced by substrate will enhance the adatom mobility at the film surface. It is generally believed the enhancement of the adatom mobility will contribue to the relaxation of compressive stress of the film resulting in an increase of the column diameter ZnO films.

4. CONCLUSIONS

In conclusion, the best quality ZnO films in terms of crystalline structure have been grown on silicon substrate at target to substrate distance 7 cm employing r.f. magnetron sputtering method from a metallic zinc target. Thermal annealing of ZnO film is performed in this condition. X-ray diffraction shows that the films are well oriented where the c-axis grows normal to the substrate plane. All the ZnO thin films deposited by rf magnetron sputtering have compressive stress. The crystallographic characteristics correlated with a direct band gap of 3,3 eV and a high resistivity of 10^{12} Ω.cm [14]. Consequently, well-oriented zinc oxide thin films with both structural properties and high resistivity could be successfully deposited by r.f magnetron sputtering.

5. REFERENCES

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