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Piezoelectric ZnO thin films by rf magnetron

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--ABSTRACT--

ZnO thin films were deposited on silicon substrate by RF magnetron sputtering using metallic zinc target. A systematic study has been made of the influence of the target to substrate distance and the RF power on the films structural properties. They exhibited a c-axis orientation of below 0.32° FWHM of X-ray rocking curves, an extremely high resistivity of 10^{12} Ωcm and an energy gap of 3.3 eV at room temperature. It was found that a substrate temperature of 100°C, very low gas pressures of $3.35x10^{-3}$ Torr in argon and oxygen mixed gas atmosphere giving to ZnO thin films a good homogeneity and a high crystallinity. Bulk acoustic wave (BAW) resonators have been fabricated by stacking different layers of Pt/ZnO/Pt on a silicon substrate which could be used for the fabrication of humidity sensors based on the quartz microbalance principle.

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

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I. INTRODUCTION

Zinc oxide is one of the most interesting II–IV compound semiconductors with a wide direct band gap of 3.3 eV [1–6]. It has been investigated extensively because of its interesting electrical, optical and piezoelectric properties making suitable for many applications such as transparent conductive films, solar cell window and MEMS waves devices [4]. The full-width at half-maximum (FWHM) of the (0 0 2) X-ray rocking curve is known to be Suppressed below about 0.5° for obtaining effective electromechanical coupling [2]. The thermal stresses were determined by using a bending-beam Thorton method [3] while thermally cycling films. ZnO has hexagonal Wurtzite structure and some properties are determined by the crystallite orientation on the substrate. For example, for piezoelectric applications, the crystallite should have the c-axis perpendicular to the substrate. According to the literature, the reactive sputtering technique has received a great interest because of its 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 properties are mainly caused by the kinetic energy of the clusters given by electric field [7-8] This energy enhances the surface migration effect and surface bonding state.

In this paper, we have developed, measured, and test the r.f. sputtred piezoelectric ZnO thin films in the bulk acoustic resonator (FBAR) devices.

II. EXPERIMENTAL

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.

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. After deposition, silver dot electrodes were evaporated on the sample ZnO/Pt/Ti/Si using an electron gun evaporation system, in order to make a metal–oxide– metal structure useful for electrical measurements. These were carried out in a vacuum chamber evacuated to approximately 10^{-3} Torr. Both the electrical conductivity and dielectric constant were measured as a function of temperature (30–250 8C) and frequency (5 Hz–13 MHz by a capacitance bridge technique), with a Hewlett-Packard LF impedance analyzer 4192 A between the silver dots and Pt electrode bottom. Temperature was measured with a Doric thermometer (Trendicator 400K/8C), and a high frequency probe station with a network analyzer (HP8753ES) were used to measure the resonant frequencies of the fabricated device

III. RESULTS AND DISCUSSIONS

3.1 Influence of target-substrate distance:

The series of samples that we conducted to optimize the deposition parameters for the influence of the target-substrate distance on the properties of ZnO layers deposited. Table 1 shows the experimental conditions for realization of this series.

Table 1: ZnO sputtering conditions

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

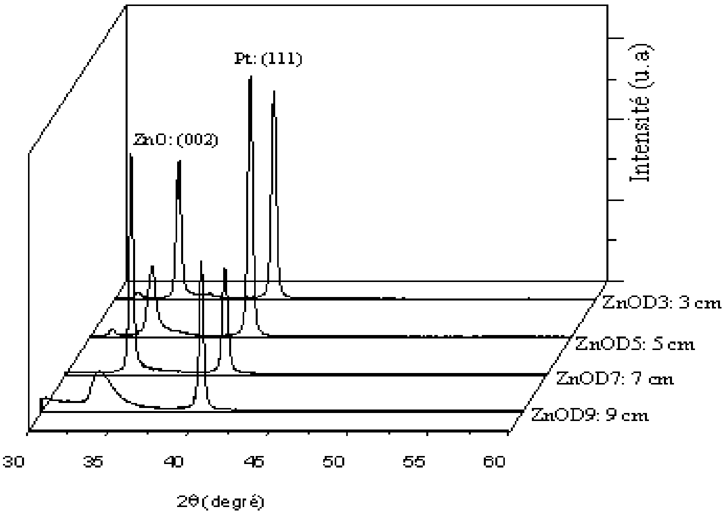

Fig.1- XRD diagram from ZnO samples deposited on silicon substrates at different target-substrate distance.

Fig.1 shows a comparison of X-ray diffraction spectrum for all samples produced at different distances target substrate.

It shows the existence of different ZnO peaks when the deposits are made to target-substrate distances less than 7 cm. Beyond that, there is the peak (002) of ZnO is present but with a poor crystallization to 9 cm.

We can say that the layers developed at a target-substrate distance of less than 7 cm are polycrystalline, although the preferred orientation of these layers are the (002) plane. Given the target substrate distance and pressure, there thermalization of the atoms during deposition.

Thus, the ideal distance to deposit ZnO thin monocrystalline, taking into account the parameters, is the target-substrate distance equal to 7 cm.

To reflect this change in the intensity of the diffraction peak and the full width at half-height of the latter, we have plotted the different target-substrate distances. Curves are shown in Fig.2. These results confirm that the sample prepared at the target-substrate distance equal to 7 cm has the best and the diffraction peak width at half height finest.

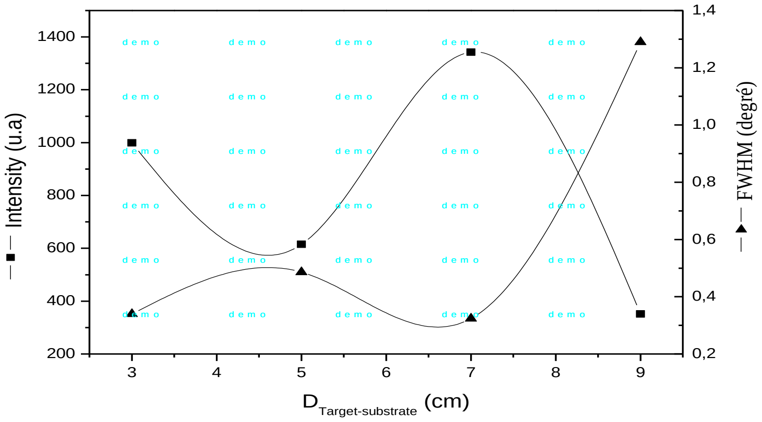

Fig.2- Evolution of the peak intensity and the width at half height of the films according to the target-substrate distance.

3.2. Effect of the power RF:

ZnO films were deposited on Si substrates with varying rf power at 100 °C substrate temperature. The surface morphologies of the grown films were investigated. Fig.3(a), (b), (c) and (d) show three-dimensional AFM images of the films grown at power P_{rf} = 50, 100, 150 and 200 W respectively.

The film grown at 200 w shows an elliptically textured shape layered parallel to surface and the small grains seen in Fig.3 (a), (b) and (c) cannot found. The sample deposited at P_{rf} = 50 W shows a smooth surface and small circular grains. However, above P_{rf} = 50 W, the surface topography was slightly roughened as shown in figure 3 (b) and (c). A reason for this is thought to be the surface damage causing by bombarding the film surface with high energy sputtered ions. In the case of ion-assisted deposition, surface damage occurs, but it is rapidly repaired when the incident ion energy is low [9]. However, when the rate of surface damage is larger than the rate of repair, a slightly rough and poor crystalline film is formed. This is in good agreement with the result for the rms surface roughness of the ZnO films. The rms surface roughness decreased as the rf power is increased. From these results, we would suggest that the power inject does not affect the surface roughening up to P_{rf} = 50 W and that the incident energy of the ions contribute only to the surface migration and the surface reaction to form a flat surface in this rf power region.

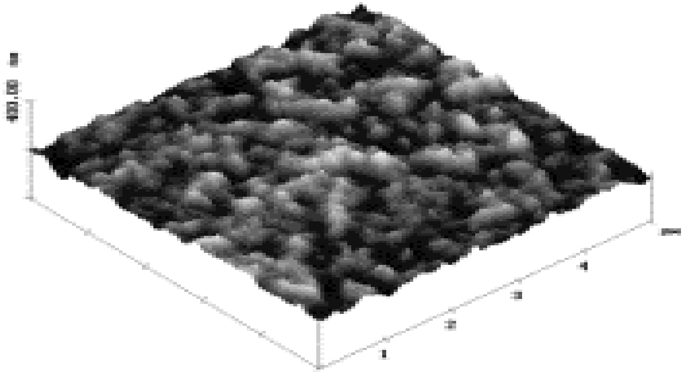

Figure 3 (a)

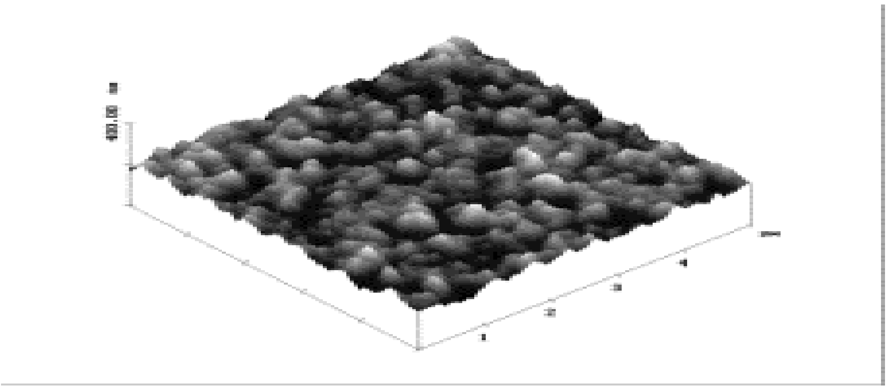

Figure 3 (b)

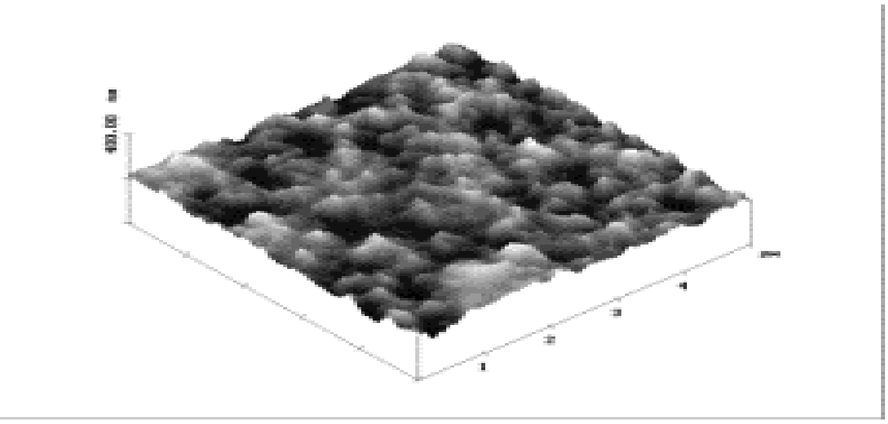

Figure 3 (c)

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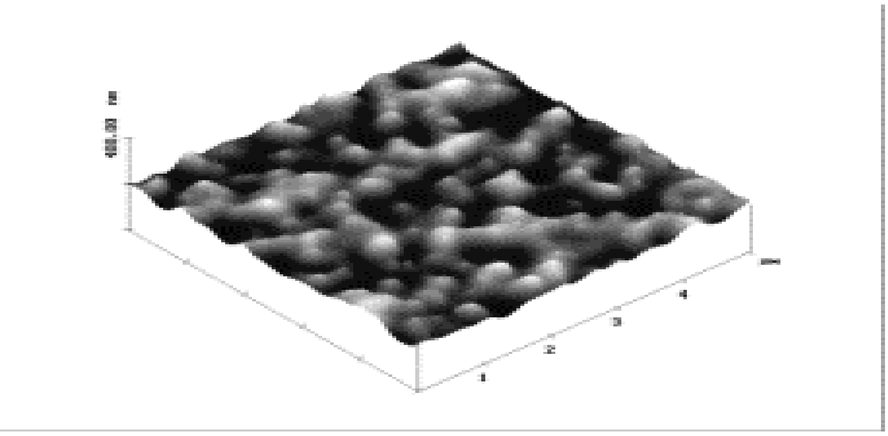

Fig 3 (d)

Fig.3- AFM image of ZnO films deposited at (a) P_{rf} = 50 W, (b) 100 w, (c) 150 W and (d) 200 W.

3.3 Crystallographic characteristics

Fig.4 shows the XRD pattern for samples deposited at several RF power (ranging from 50 to 200 W). The peak at about 34° corresponds to the diffraction from the (0 0 2) plane of the ZnO. No other peak could exhibit a preferred c-axis orientation. The intensity of the (0 0 2) peak decreased as the power inject is increased.

The film deposited at P_i = 50W show a (0 0 2) peak a high intensity. This can be explained by the nucleation kinetics and the film growth which are influenced by the interactions between the substrate and the incoming energetic atoms on the ZnO films crystallinity. It is noticed however that Si substrate makes it possible to have crystalline structure films deposited near to pure zinc oxide. However at higher power inject, the (0 0 2) peak intensity decreased which indicates that the crystallinity of the films is deteriorated.

Earlier reports indicate that the films formed at a power inject of 50W had a preferred orientation along the (0 0 2) crystal plane. Again a preferred orientation along the (0 0 2) crystal plane at 150–200W in DC magnetron sputtering films [10]. The variation in the results may be due to differences in the deposition methods.

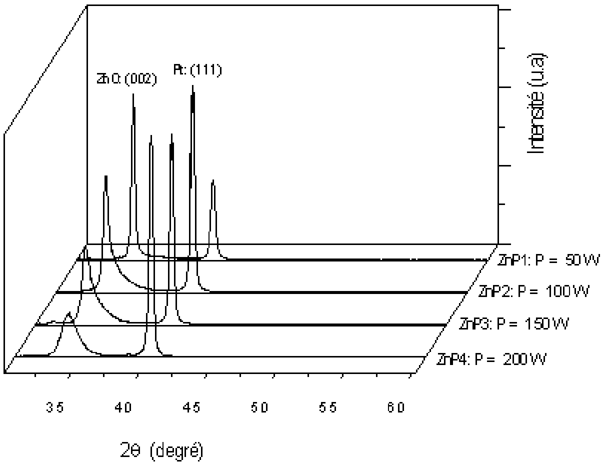

Fig.4- XRD diagram from ZnO samples deposited on silicon substrates at different rf power

3.4. Resistivity for ZnO/Si

Measurements of resistivity were made at 10^3KHz using a Hewlett-Packard427 5A LCR Meter. Fig.5 shows the effect of RF power on the resistivity of ZnO films. The resistivity decreases with increasing of RF power. This phenomenon suggests that atoms produced with larger driving force to migrate to more suitable lattice and result in low perfect crystals. The thin film is known to have higher resistivity as the structure is more crystalline. The sample deposited at 50Wpresent a resistivity of $10^{12}\Omega$cm. [11].

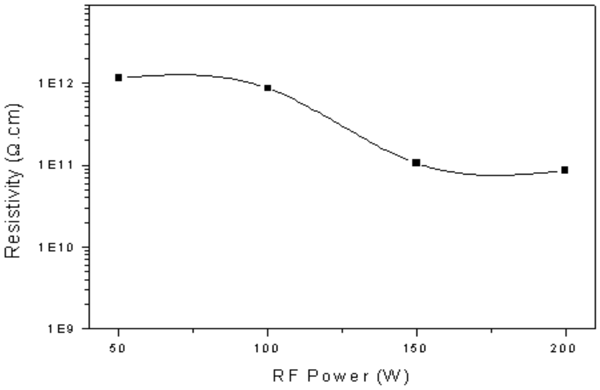

Fig.5- Resistivity of ZnO films as a function of the Effect of rf power.

3.5. Electrical admittance for ZnO/Si

Fig.6 shows the variation of the real and imaginary parts of the admittance electrical function of frequency obtained on one of our ZnO layers.

We are seeing a greater or lesser variation of the electrical input admittance of the resonator made and the appearance of oscillations superimposed on the overall envelope. These oscillations are due to the propagation medium. They reflect the fluctuation bet real and imaginary admittance measured.

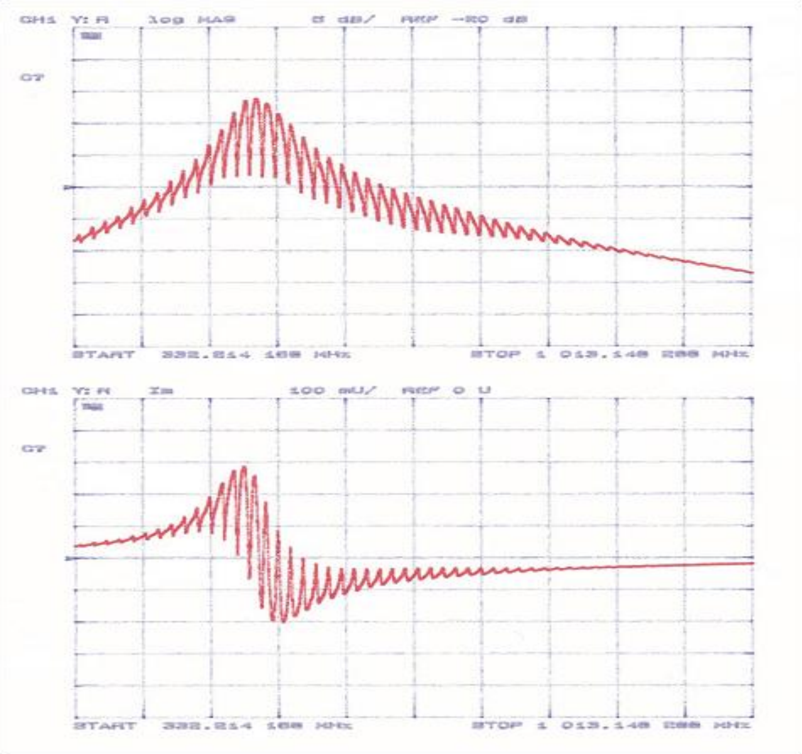

Fig.6- Electrical admittance of the resonator ZnO.

Indeed, Fig.7 proves there recombination waves propagating in the medium of propagation are the silicon substrate. This leads to the change in acoustic impedance returned to the surface of the ZnO layer. These oscillations appear with a frequency of 11 MHz.

We also measured the evolution of the reflection coefficient as a function of frequency on a Smith chart. We observe in Figure 8, the presence of piezoelectric activity in ZnO. The appearance of the resistive effect and change the capacitive effect clearly reflect the conversion of electrical energy into mechanical energy. Thus, the maximum of the real part of the admittance, which represents the frequency f_S series resonance of the structure, was observed near the fundamental mode at 520 MHz and the minimum of the susceptance at resonance, representing the parallel resonance frequency f_p the structure is observed at 537 MHz.

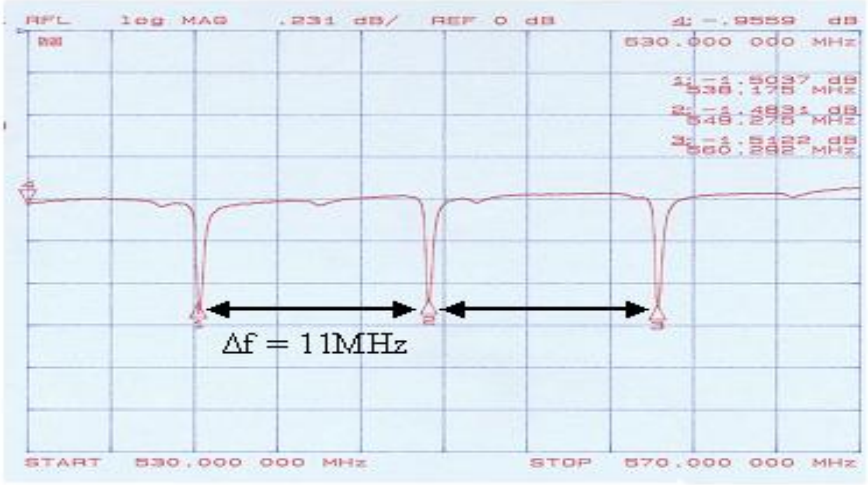

Fig.7- Display oscillations in the substrate.

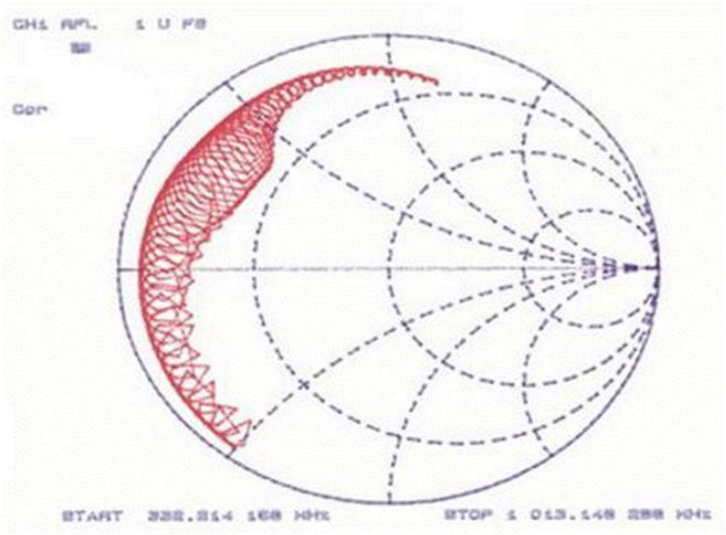

Fig.8- Visualization of the piezoelectric activity of the Smith chart.

IV. CONCLUSION

Piezoelectric ZnO film was deposited using RF magnetron sputtering to fabricate an FBAR device. 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. 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 [11]. Consequently, well-oriented zinc oxide thin films with both structural properties and high resistivity could be successfully deposited by r.f magnetron sputtering. The piezoelectric activity was observed of the ZnO film. The series and parallel resonance frequencies of the FBAR device appeared at 520 and 537 MHz, respectively, which represent 92% of the values for a ZnO FBAR of the same thickness without a mass loading effect.

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