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Deposition of ZnO Layers Using Planar Reactive Magnetron System

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Introduction

Although ZnO thin films have been extensively investigated over the last 30 years they have received a rapidly growing interest due to their wide range of scientific and technological applications, such as thin piezoelectric film surface acoustic wave (SAW) devices, transparent conductive coatings, thin film gas sensors, optical waveguides and laser deflectors, light modulators and optical sensors. The optical gap of ZnO film is 3.3 eV, so these films are transparent in a large wavelength range. Trivalent donor impurity doped ZnO films are *n*-type semiconductor with a low electrical resistivity. Otherwise, doping of acceptor impurities increases specific resistance of ZnO. Crystals of ZnO belong to a group of the hexagonal wurtzite and highly oriented polycrystalline ZnO films have strong piezoelectric features [1–3].

Unique ZnO film features depend on technology of growth process. Chemical vapor deposition, spray pyrolysis, molecular beam deposition, pulsed laser ablation and different modifications of sputtering methods are used for deposition of thin ZnO films [2, 3].

Investigation of highly oriented polycrystalline ZnO films growth was the main aim of this work. These films are suitable for manufacturing thin film SAW interdigital transducers (IDT) for excitation of SAW in non-piezoelectric materials. The analysis of SAW propagation parameters (velocity, attenuation) can be used for non-destructive investigation of thin non-piezoelectric materials properties because the main part of SAW energy is concentrated in thin surface layer, thickness of which is approximately equal to SAW wave length [4].

Experimental

The main requirements for thin polycrystalline piezoelectric ZnO films for manufacturing SAW transducers are [2]:

1. Polycrystalline ZnO films have to be optically transparent for acousto-optic applications and have a thick columnar structure.
2. The *c*-axis of ZnO crystallites have to be highly oriented perpendicular to a substrate surface. X-ray examination of piezoelectric films is a major tool for determining the uniformity of crystalline structure.
3. High electrical resistance of ZnO films is necessary in order to reduce SAW attenuation due to interaction between conductivity electrons and supporting SAW periodical electric field. Single-crystal piezoelectric semiconductors with resistivity from 10^3 to 10^6 $\Omega\cdot$cm are required for an acoustic amplifier.
4. The thick ZnO layers (thickness approximately is equal to half wavelength of SAW) commonly are required for effective SAW excitation in non-piezoelectric materials. For this reason high deposition rate technologies are preferable.

The deposition methods of magnetron sputtering in direct current and radiofrequency regimes using metallic Zn and ZnO targets in oxygen and oxygen-argon gas mixture atmosphere were experimentally investigated in our work.

ZnO layers were deposited on the glass K-8 substrates. ZnO layer deposition rate, electrical resistivity, crystalline structure, and piezoelectric features of deposited ZnO layers were analyzed.

The crystalline structure and orientation of deposited ZnO layers was investigated using X-ray diffraction (XRD).

The sheet resistance of ZnO layers was measured by the four probe technique.

Piezoelectric properties of deposited ZnO films were estimated using comparison between theoretically calculated SAW delay line characteristics and experimental measurement results.

Results and discussion

Low ZnO deposition rate (up to 1 μm/hour) was the main disadvantage of radio frequency ZnO sputtering system. In the case of ceramic ZnO targets increasing the cathode current is limited by low ZnO ceramic thermal conductivity. Overheating of ZnO ceramic cathode leads to its physical destruction. The higher DC discharge current density can be reached in the case of water-cooled metallic Zn targets. Reactive gas mixture (argon and oxygen) in sputtering vacuum chamber must be used in this case. The increase of argon concentration in gas mixture increases ZnO deposition rate in the case of the same current density but the electrical resistance of ZnO layers decreases.

The best result in piezoelectric ZnO layers deposition was achieved using planar reactive magnetron system with metallic Zn target (Fig. 1).

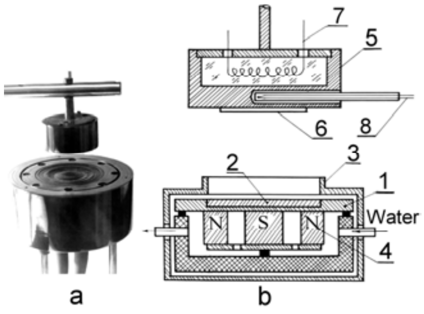


Fig.1. View (a) and construction (b) of planar magnetron system: 1 – cathode, 2 – metallic Zn, 3 – anode-shield, 4 – permanent magnet, 5 – substrate holder, 6 – substrate, 7 – heating coil, 8 – thermocouple

Deposition in oxidizing ambient (Ar and O_2 gas mixture) from metallic zinc cathode (target) in our case was preferable. The following advantages of process were experimentally proved:

1. Heat conductivity of metallic zinc is higher than ZnO ceramic. It results in realizing larger cathode current density and higher deposition rate.

2. Power applied circuit in direct current magnetron system is simpler (less expensive).

3. Cathodes from high purity metallic zinc are easily produced. Controlled amount of desirable impurities can be introduced during cathode producing process.

Sputtering in magnetron deposition system can be realized in higher pressure range with respect to diode or triode sputtering system. It additionally raises the deposition rate. The component of magnetic field directed parallelly to substrate enables to reduce the bombardment of substrate and coating by secondary electrons in magnetron sputtering system. It results in reducing radiation defects density of growing layer.

Magnetron target with permanent magnets system (Fig. 1a, Fig. 1b) was used there. Tangential magnetic field in cathode sputtering region was about 0.08 T, plasma ring internal diameter – 70 mm, its width – about 15 mm. Current-voltage characteristics of used magnetron-substrate holder system is presented in Fig 2. Substrate temperature kinetics during deposition for different discharge power values (without additional substrate-holder heating) is shown in Fig 3. The strong dependence of microstructure and stoichiometry of deposited layers on substrate temperature during the process was observed in all experiments. For better reproducibility of results, holder with substrate was heated before coating to desirable temperature using build-in substrate holder heater (Fig. 1b). The temperature of substrate was controlled by thermopile and stabilized during all process of deposition. Desirable pressure (and gas composition) in the limits presented in Fig. 4 were stabilized before beginning of ZnO deposition.

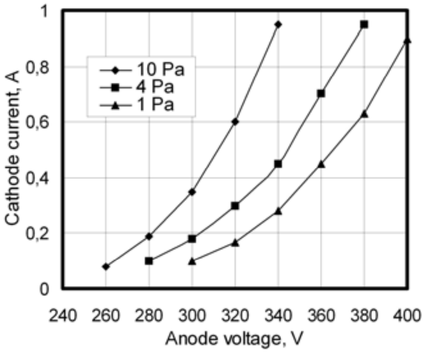


Fig. 2. Current-voltage characteristics of magnetron discharge at oxygen pressure 1 Pa, 4 Pa, and 10 Pa

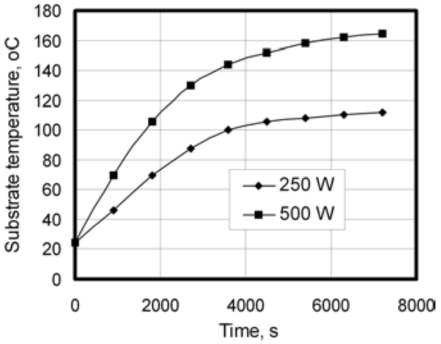


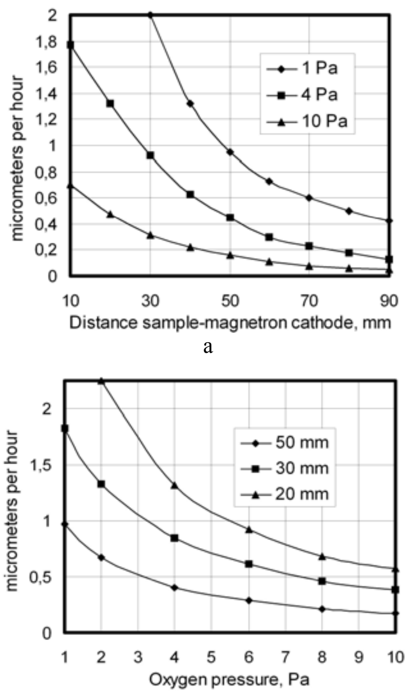
Fig. 3. Substrate temperature versus time in the case of discharge in 1 Pa of oxygen ambient for distance between magnetron and substrate of 30 mm and discharge power of 250 W and 500 W respectively

Dependences of deposition rate on the distance between target (cathode) and substrate and partial pressure of O_2 in vacuum chamber during coating are presented in Fig. 4a, Fig. 4b.

ZnO layers were deposited on glass K-8 substrate in our experiment. The deposition of thick piezoelectric ZnO layer (thickness about 20 μm) with the maximum deposition rate was proposed as the object. No essential difference in stoichiometry of ZnO films was observed for all used substrates in the case of the same gas pressure and deposition rate.

The higher rate of ZnO growth was obtained in magnetron sputtering system using O_2 and Ar gases mixture ambient. Unfortunately, increasing of argon partial pressure leads to decrease of layers electrical resistivity. The possibility of obtaining the high resistivity ZnO films was realized in the case of pure oxygen plasma use. Increasing of Ar partial pressure leads to increase of deposition rate but in our case it was unacceptable due to low resistance of ZnO layers. High resistance coating was produced using magnetron discharge in pure oxygen ambient.

In the case of low deposition rate (up to 1 μm per hour) and without additional substrate heating disoriented polycrystalline non-piezoelectric ZnO films were



a

b

Fig. 4. Dependences of ZnO deposition rate for discharge power 100 W: a – on the distance between substrate and magnetron cathode for oxygen pressure 1 Pa, 4 Pa, and 10 Pa; b – on oxygen pressure for distances between substrate and magnetron cathode 20, 30, and 50 mm

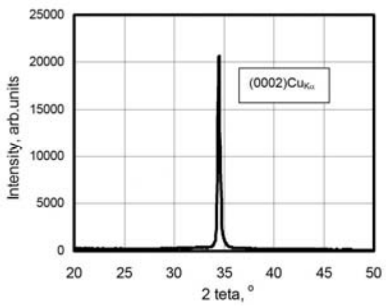


Fig. 5. X-ray diffraction pattern for 20 μm thick ZnO film on K-8 glass

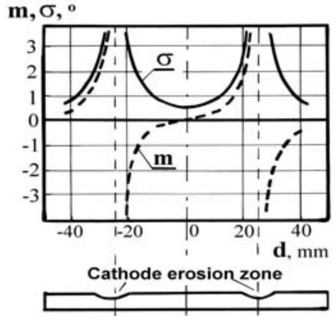


Fig. 6. Results of XRD investigation of ZnO films structure on substrate surface: d – distance from substrate centre; σ – standard deviation of *c*-axis of ZnO crystallites; m – declination of dominant *c*-axis of ZnO crystallites

deposited. The optical transparency of such films was low. Experiments at different substrate temperatures and at different deposition rates demonstrated the increase of crystallites orientation degree (examined by XRD) in the case of substrate temperatures increase (up to 450 °C) and deposition rate increase. Temperature dependences of XRD results were about the same for deposing ZnO on all used substrates. The optical transparent ZnO layers in the case of the thickness up to 20 μm were produced for deposition rates of 10 μm/hour and more. The substrate temperature during this process exceeded 400 °C. Optimal oxygen pressure in vacuum chamber was about 1 Pa.

Results of XRD investigation particularly presented in Fig. 5 and Fig. 6 demonstrated high oriented texture of ZnO film with normal *c*-axis to substrate surface.

Standard deviation σ of *c*-axis orientation depends on distance between the substrate centre and analyzed point. This dependence is schematically presented in Fig. 6. The largest declination of dominant crystallites orientation from direction of substrate normal *m* was observed in deposited film areas located against cathode erosion zone. Correlation between structure of deposited ZnO film and the flux of high-energy neutral atoms was demonstrated in [6]. Maximum declination angle is in the region of substrate where the largest flux of high-energy atoms during deposition reaches the substrate. (Fig. 6). Consequently, high oriented polycrystalline ZnO film was deposited not on the whole substrate surface. The shape of random oriented layer region and its area depends on cathode erosion zone shape and magnetic field configuration.

The electrical resistance of piezoelectric layer is very important factor in applications of ZnO films for SAW interdigital transducers. It is desirable to obtain resistivity of ZnO as high as possible.

The resistivity of deposited ZnO layer was 10^4 – 10^6 Ω·cm in dependence on deposition conditions. Annealing of deposited layers in atmosphere increased the resistivity up to 10^7 Ω·cm. Additional increasing in approximately two order of resistivity value was observed in the case of using for ZnO deposition Zn cathode doped by 0.5 % mass of Cu. Incorporation of Cu can take place at interstitials as well as substitutional sites to increase resistivity of films. Doped Cu atoms in ZnO involve their substitution for Zn atoms; they act as acceptors that compensate the donors (excess of Zn atoms).

Piezoelectric properties of ZnO layers were investigated using acoustic delay lines (Fig. 7) [4]. Each of them consists of two SAW interdigital transducers, formed on non-piezoelectric substrate (glass K-8) using deposited piezoelectric ZnO films on IDT region through mask. Thin (about 300 nm) film of aluminium was evaporated on glass and arrays of IDT electrodes were formed using convenient photolithography technology. Each of them consists of 24 pairs of fingers. Period of grating was 37.5 μm, aperture – 2.5 mm, distance between IDT midpoints – 13 mm.

After producing IDT electrodes, substrate (glass) was placed into vacuum chamber and ZnO layer was deposited on transducers through the mask. Optimal thickness of piezoelectric layer was about 20 μm.

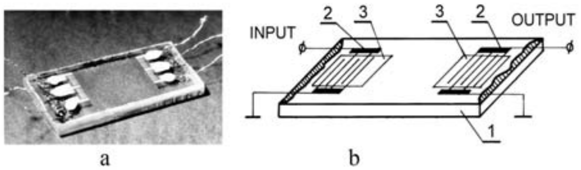


Fig. 7. View (a) and schematic diagram (b) of SAW delay lines:
1 – glass K-8 substrate, 2 – SAW transducers electrodes,
3 – piezoelectric ZnO film

Theoretic estimations of such IDT characteristics are [5]: radiation resistance on 75 MHz resonance frequency – 50 Ω, static capacity – 5.6 pF, wavelength of SAW in K-8 glass (non-coated by ZnO area) – 45.3 µm. Minimal loss in such delay line in the case of using matching inductances at IDT circuits (theoretical estimation) is 12 dB. Experimental observed loss was 14 – 18 dB.

Piezoelectric properties of deposited ZnO films were estimated using comparison between SAW delay line theoretical characteristics and results of experimental measurements. In the case of the best fitting experimental and calculation results, based on single-crystal ZnO properties, the optimal piezoelectric ZnO layers for SAW IDT deposition conditions were determined.

Conclusions

In this research focus on two points of ZnO film characteristics: high resistivity and *c*-axis (002) orientation was directed. Film deposition by RF magnetron sputtering system using metallic Zn target with 0.5 of % Cu and 400 °C temperature of substrate guarantee high resistivity (up to 10^9 Ω·cm) and strong (002) preferred orientation.

These low conductivity thick Cu doped ZnO films have potential to be used as a piezoelectric film for SAW interdigital transducers.

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Ištirtos ZnO sluoksnių savybės jiems auginti naudojant aukštojo dažnio ir nuolatinės srovės magnetronines joninio dulkinimo iš ZnO keramikos ir metalinio Zn taikinių sistemas, dirbančias deguonies ir deguonies-argono aplinkose. Plėvelės buvo nusodinamos ant K-8 stiklo padėklo. Nustatyta Cu priemaišų bei terminio ZnO atkaitinimo ore įtaka ZnO sluoksnių savitajai varžai. Parinkti optimalūs pjezoelektrinių polikristalinių ZnO sluoksnių auginimo, naudojant planarinį magnetroną, režimai. Naudojant užaugintus polikristalinius ZnO sluoksnius pagamintos paviršinių akustinių bangų vėlinimo linijos, kurių eksperimentiškai išmatuoti parametrai artimi teoriniams, apskaičiuotiems naudojant monokristalinio ZnO parametrus. Il. 7, bibl. 5 (anglų kalba; santraukos lietuvių, anglų ir rusų k.).

J. Daugėla, S. Joneliūnas, A. Jotautis, R. Naujokaitis, E. Klovaitė. Deposition of ZnO Layers Using Planar Reactive Magnetron System // Electronics and Electrical Engineering. – Kaunas: Technologija, 2005. – No 7(63). – P. 70-73.

Properties of ZnO films deposited using radiofrequency and direct current magnetron sputtering systems with ZnO and metallic Zn targets in oxygen and oxygen-argon gas mixture atmosphere on glass K-8 substrates were experimentally investigated. The influence of Cu impurities and ZnO layers post deposition thermal annealing is estimated. The optimal growth technological conditions of highly oriented polycrystalline piezoelectric ZnO films using planar reactive magnetron system are defined. The parameters of formed SAW delay lines were approximately equal of theoretically calculated for monocrystalline ZnO. Ill. 7, bibl. 5 (in English; summaries in Lithuanian, English and Russian).

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Исследованы характеристики слоев ZnO, для осаждения которых использовалась магнетронная система ионного распыления катодов из ZnO керамики и металлического Zn в режимах постоянного и высокочастотного тока. Пленки осаждались на подложки из стекла К-8. Определено влияние примесей Cu и термического отжига в атмосфере слоев ZnO на их сопротивление. Определены оптимальные режимы магнетронной системы выращивания поликристаллических слоев ZnO, обладающих пьезоэлектрическими свойствами. Параметры изготовленных линий задержки на ПАВ хорошо согласуются с теоретически рассчитанными значениями для монокристаллического ZnO. Ил. 7, библ. 5 (на английском языке; рефераты на литовском, английском и русском яз.)