**Deposition of aluminum doped ZnO as electrode for transparent ZnO/Glass surface acoustic wave devices**

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### Abstract: Unlike metal electrodes such as Al and Au, aluminum doped zinc oxide (AZO) with high conductivity and transparency can be used as transparent electrodes for surface acoustic wave (SAW) resonators, and thus realize fully transparent and invisible ZnO SAW devices on glass. This paper reports the fabrication of transparent SAW resonators using AZO as the transparent electrode and investigation of effects of deposition parameters on the crystal structures of the AZO thin films deposited by magnetron sputtering. Results show that a low sputtering pressure and an optimal sputtering power are beneficial for the deposition of (0002) orientation of AZO. The optimal deposition parameters are found to be: deposition pressure of 0.3 Pa, sputtering power of 300 W, substrate temperature of 200 °C. The fabricated transparent SAW devices have different wavelengths (from 16 to 32 μm) and all the devices exhibit two types of wave modes: Rayleigh and Sezawa waves. Compared with Sezawa wave, the Rayleigh wave has a large signal amplitude up to 25 dB. In addition, as the wavelength increases, the resonant frequencies of both the Rayleigh and Sezawa waves increase whereas their phase velocities decrease. The transparent SAW devices have also demonstrated their ability to induce a strong acoustic streaming in a water droplet with a streaming velocity up to 2.27 cm/s. This research opens a door for further exploration of the SAW devices in transparent electronics.

**Keywords:** Transparent SAW, AZO interdigitated electrode, glass substrate

1. **Introduction**

Transparent electronics (also referred as invisible electronics) have attracted great attention in recent years due to their great potential to make significant commercial impact in variety of areas. Great effort has been made to develop various transparent electronic devices and systems such as transparent thin-film transistors, photovoltaic cells, electronic circuits, displays, sensors, solar cells, and electro-optic devices etc. [[1-4](#_ENREF_1)]. However, there are few studies on the transparent surface acoustic wave (SAW) devices.

SAW devices are one of the building blocks of electronic devices with widespread applications in communications as filters, frequency duplexers and radio frequency identification (RFID) etc. [[5](#_ENREF_5), [6](#_ENREF_6)]. Recently SAW devices have also found tremendous applications in biochemical sensing, drug delivery, life science and medical research etc. such as (1) micro-sensorsto measure physical parameters and detect biochemical substances; and (2) microactuators for microfluidicsand lab-on-a-chip[[7](#_ENREF_7)]. If SAW devices can be made to be transparent on glass substrates using the transparent electrodes, they could be used for various new applications. For examples, they can be used as invisible wireless RFIDs and SAW sensor arrays on windows of cars and offices, and on screens of eyeglasses, helmets and other electronic gadgets to monitor environmental conditions such as pressure, temperature and ultra-violet (UV)-light, as well as to detect biochemical molecules associated with air pollution and biological attachment. Most of the current SAW devices are typically made on piezoelectric bulk materials or thin films deposited on solid substrates such as Si and sapphire, consisting of a pair of interdigitated electrodes (IDT) which are normally made of metals such as Al and Au. Therefore, the so-made SAW devices are opaque and visible.

If the IDTs of SAW devices are made of transparent electrode materials, the SAW devices can be fully transparent on a piezoelectric film coated glass substrate. As both glass substrate and piezoelectric film (e.g. ZnO or AlN film) are intrinsically transparent, the key focus of this study is to develop transparent IDT electrodes. There are several possible electrode materials that could be used and the most commonly used material is Sn-doped indium oxide (ITO). ITO has been extensively studied for applications in flat panel display devices because it has a low resistivity, high optical transmittance, and chemical stability. However, ITO has several disadvantages including toxicity, high price and instability in hydrogen plasma [8]. Alternatives to ITO have recently been reported, which include carbon nanotubes [9] and graphene [10]. However, the SAW application using these carbon-based materials is probably limited by their low conductivity [11], as the sheet resistances for IDTs of SAW devices should be lower than 65 Ω/Sq [12]. Even though the sheet resistances of the carbon-based materials such as graphene could be decreased by post processing such as multilayer transfer and doping, but the process is complicated. Other examples include silver nanowires (AgNWs) based electrodes [13], which can achieve a sheet resistances less than 10Ω/Sq with 90% optical transmission because of the high conductivity of the silver. However, practically it is not easy to pattern the AgNWs IDTs with the finger width less than 10 μm. Furthermore, the AgNWs are easily damaged and removed when they are immerged into acetone and DI water which are the key fabrication process of IDTs’ patterning. Al-doped ZnO(AZO) is another good alternative to ITO due to its high electrical conductivity and optical transmittance, and transparency to most of visible light owing to the wide energy band gap of 3.3 eV. In addition, the AZO film can be fabricated by magnetron sputtering which is a low cost and good reproducibility method [14]. In 2013, Zhou et al reported the transparent SAW devices with AZO electrodes [12]. In that communication, transparent SAW devices were proposed, and the transparent SAW devices generated Rayleigh mode SAWs, which has been successfully used as temperature sensors. Furthermore It was demonstrated that the sheet resistances of IDTs should be less than 65Ω/Sq [12] in order to achieve good performance of SAW devices. In 2014, Chen et al reported flexible and transparent SAW devices with ITO/ZnO/flexible glass structure using conventional ITO electrodes [15]. The flexible and transparent SAW sensors demonstrated good capability to detect strains in a very wide range from -3000 to 3000 με, with a sensitivity of ~34.7 Hz με-1. In 2016, DeMiguel-Ramos et al reported the transparent bulk acoustic wave resonators (FBARs) with a resonance frequency of 1800 MHz, electro-mechanical coupling coefficient *K*2 of 4% and transparency of 75% [16].

This paper is focused on the effects of deposition parameters on the crystal structures of the AZO thin films, which are explored as the transparent electrode for SAW devices. The transparent SAW devices have been fabricated using AZO transparent electrodes. Microfluidic application was demonstrated using the fabricated transparent SAW devices.

1. **Experimental**

AZO film was deposited using an in-house developed direct-current (DC) magnetron sputtering system with an AZO target (ZnO 98 wt.%: Al2O3 2 wt.%,) with a diameter of 50 mm. The base pressure of chamber was Pa before deposition. The distance between the target and substrate was fixed at 70 mm. The substrate holder (100 mm in diameter) was rotated during the deposition to achieve films with a good uniformity. Effects of deposition conditions such as the pressure and DC power on the properties of the AZO films were investigated. Table 1 summarizes the deposition parameters.

Crystalline structure and crystal orientation of the films were analyzed by X-ray diffraction (XRD-6000, JAPAN) using Cu- radiation and a scanned range of 2θ =20°~70°. The degree of c-axis crystallization was examined by the full-width at half maximum (FWHM) of the AZO (0002) diffraction peak. Crystallite grain sizes were calculated from the Debye–Scherrer formula: , where *K* is the shape factor of the average crystallite with a value of 0.94, *λ* the X-ray wavelength (1.5405 Å for Cu target), *β* the FWHM in radians, *θ* the Bragg angle, and *D* the mean crystallite gain size normal to diffracting planes. For cross-sectional structural analysis, a scanning electron microscope (SEM, S4800, HITACHI Company, JAPAN) was used.The resistivity of the AZO layers was measured using the Van de Paul Hall effect measurement. Optical transmittance of the AZO films was measured using a UV/visible spectrometer (UV756).

SAW devices were made on 2-inch Corning glass 2318 substrates of 1.1 mm thickness, with ZnO thin films as the piezoelectric layer which were deposited using a DC reactive magnetron sputtering system. The optimal deposition conditions of the ZnO film were as follows: substrate temperature of 100 °C, deposition pressure of 2 Pa, sputtering power of 200 W, O2/Ar gas of mixture 50/100 sccm and bias voltage of -75 V [[12](#_ENREF_13)]. The obtained thickness of ZnO film is 3 μm. After deposition of the ZnO film on glass substrate, the IDT electrodes of AZO were formed using the standard lift-off process. The wafer was patterned by photolithography using a positive photoresist (RZJ304) to form IDT electrode patterns. The patterned wafer was then put into the sputtering chamber for deposition of the AZO layer. Once completed, the wafer was immersed into acetone to remove photoresist and unwanted AZO to obtain the IDT electrodes. The distance between the two IDT transducers is 20 λ, where the wavelength, λ, is determined by the IDT pitch. The transmission (S21) characteristics of the SAW devices were measured using an Agilent E5071C network analyzer.

1. **Results and discussion**

### 3.1 Deposition pressure effect

Figure 1(a) shows the XRD patterns of the AZO films deposited under different deposition pressure values. All the AZO films show a dominant XRD peak at 2θ = 34.4°, which corresponds to (0002) crystal orientation of the AZO. They are 99.85% close to that of the standard ZnO crystal (34.45°), independent of the Ar gas pressure. No Al2O3 phase was detected from the XRD patterns, indicating that all the Al atoms replace the zinc substitutionally in the hexagonal lattice, or Al segregate to the non-crystalline region in the grain boundary[[17](#_ENREF_14)]. Based on Fig.1, with the increase of pressure, the intensity of the (0002) peak of AZO decreases. Moreover the FWHM values of the (0002) peak of AZO increase, and the grain sizes decrease with the increase of the gas pressure, which is consistent with the literature [[18](#_ENREF_15)]. The decreased intensity of the XRD indicates relatively poor quality of AZO films was obtained at higher deposition pressure. As increasing the deposition pressure leads to the decrease of kinetic energy of sputtered atoms and to a low surface mobility of the condensing species, especially at low deposition temperatures, the (0002) crystallization decreases when the pressure is increased. The lowest deposition pressure used is 0.3 Pa, and below this value, the glow discharge will become extinguished.

Figure 2 shows the resistivity, carrier concentration and Hall mobility of the AZO films deposited at different deposition pressures. As the pressure is increased,the resistivity of the AZO film increases, while the carrier concentration and Hall mobility of the AZO films decreases, which is consistent with those in literature [[19](#_ENREF_16)]. This demonstrates that with the increase of pressure, the quality of AZO film decreases which coincides with the XRD results. This phenomenon can be explained using the basic theories of adatom mobility and scattering effects. At higher deposition pressure, the probability of scattering effect increases and more particles could lose their kinetic energy by collisions before reaching the substrate, thus resulting in a lower adatom surface mobility. Generally, a higher adatom mobility promotes the growth of the AZO thin film with a c-axis orientation, because if the particles have more mobility they are more likely to find the low energy binding sites, thus enhance the crystal growth along its preferred orientation. Furthermore, at a high deposition pressure, the crystalline grains of the AZO decrease, thus increasing the intergranular defects and scattering effect, leading to a higher resistivity of the AZO films. The lowest resistivity of the AZO layers measured using the Van de Paul Hall measurement is about 4x10-4 Ω·cm, and the Hall mobility is 15.6 cm2/V·S-1, similar to most reported values [[18](#_ENREF_15)].

Optical transmittance of the AZO films measured using the UV/visible spectrometer is shown in Fig. 3. The results show that the transparency of each AZO film is close to 90%, except the film deposited with a deposition pressure of 1.5 Pa. The high transparency demonstrates good quality of the AZO films.

### 3.2 Deposition power effect

Figure 4 shows SEM micrographs of the cross-sectional structure of the AZO films deposited at different deposition powers with the deposition pressure of 0.3 Pa, temperature of 200 °C and deposition time of 0.5 h. Results show that the AZO crystal structures are perpendicular to the substrate and all the AZO films exhibit a typical (0002) oriented columnar structure. As the power is increased from 100 to 300 W, the columnar structures become more neatly arranged and compact. A possible reason is that a reasonably high sputtering power not only increase the sputtering yield (see Fig. 4(f) of deposition speed), but also increases the incident particle kinetic energy which enhances the mobility of the atoms deposited and the migration distance, thus is beneficial for the growth of (0002) oriented crystallites. However, when the power exceeds 300 W, the structures became coarser and non-columnar as shown in Fig. 4(e). This is because while the sputtering power is 400 W, the deposition rate is as high as 38.5 nm/s, and atoms might not have sufficient time to rearrange on the surface before the next atoms are deposited, thus leading to a porous crystal structure. Furthermore, the incident atoms have very high energy, which can damage the surface of the newly formed AZO layer, thus destroying the (0002) crystal orientation. Figure 4(f) shows the deposition rate as a function of deposition power, indicating it is linearly correlated to the deposition power.

Figure 5 shows the dependence of the FWHM of (0002) oriented AZO films. The results show that the sample deposited with a power of 300 W has a minimum FWHM of 0.304° with an average grain size of 29 nm, implying this is an optimal power for the best crystal quality, which is consistent with the SEM investigation.

From the above analysis, the optimal deposition conditions of AZO film were obtained: deposition pressure of 0.3 Pa; sputtering power of 300 W, and substrate temperature of 200 °C. With a deposition time of 0.5 h, a film thickness of 750 nm could be obtained.

**3.3 Transparent SAW device results and microfluidic application**

Figure 6(a) is the schematic structure of the fabricated AZO/ZnO/Glass SAW devices. Figure 6(b) is an SEM picture of the AZO layer on top of ZnO layer for the device, showing a clear columnar structure with large grain size for both the ZnO and AZO film, and the preferential c-axis orientation which is critical for high transparency and good performance SAW devices. Optical transmittance for the layered AZO/ZnO films on glasses was measured using a UV/visible spectrometer and it is over 80%. Figure 6(c) shows the fabricated SAW devices on a glass wafer, showing a good transparency, by revealing clearly the background features. Figure 6(d) is a zoomed-in SAW device with the AZO electrode.

The transmission (S21) and the reflection (S11) characteristics of the transparent SAW devices are shown in Fig. 7. The first mode resonant frequency, *f*1, was found to be 154.9, 129, and 85 MHz, respectively, and the second mode resonant frequency, *f*2, from 320, 272 and to 175 MHz as the wavelength of the IDTs was increased from 16 to 32 μm. A large signal amplitude up to about 25 dB was obtained for the first wave mode. These results clearly demonstrated that the transparent ZnO films have good piezoelectric effects, and the AZO layer functions well as the transparent electrodes. The amplitudes of the first mode resonance for all the devices are larger than the second mode resonance, implying the first mode resonance of the devices can be easily excited.

To verify the vibration patterns of these two modes, we have performed simulation using the finite element analysis (FEA) with commercial COMSOL 5.1 software in a two-dimensional (2D) piezo plane strain mode. The IDTs are periodic in nature, alternatively consisting of positive and negative potentials. Thus, one period of the electrode is sufficient to illustrate the performance of the SAW resonator. In this work, the experimental results of the transparent device (see Fig. 7(c)) with a wavelength of 32 μm were chosen for comparison with the simulation results. A model of an ZnO film with a thickness of 3 µm on a 60-µm thick glass substrate (about 2λ, thick enough for simulation, as the acoustic wave only penetrates into the substrate about one wavelength depth) was simulated with a fixed bottom boundary condition as shown in Fig. 8(a). A free and zero charge/symmetry boundary condition was assigned to the top surface of the piezoelectric ZnO layer. An electric potential with value of 1 V was assigned to the 750 nm thick AZO electrode, while the other AZO electrode was assigned to be ground. The boundary between the ZnO and glass was assigned to be free and continuous. The two sides of the ZnO and glass substrate were assigned to be periodical boundary conditions. The material constants were extracted from the material library of COMSOL.

The simulation results are shown in Fig. 8(b). Clearly there are two large resonant peaks in the simulated frequency range which correspond to two SAW wave modes. The first peak is at 88.6 MHz, which corresponds to the experimental results of 85.0 MHz. Surface displacement, vibration and deformation of this mode are shown in Fig. 8(c), showing that the first peak is the Rayleigh wave-mode. Whereas the frequency of the second mode is 181.2 MHz from the simulation and the particle vibration analysis as shown in Fig. 8(c), confirming that the second wave mode is the Sezawa mode.

The phase velocity for the Rayleigh wave in an ideal (0002) ZnO layer is ~2650 m/s[[20](#_ENREF_17)], depending on the crystal quality and deposition method used. The phase velocity for the Rayleigh wave in glass is 3200 m/s[[21](#_ENREF_18)], which is larger than that of ZnO film. This is a layered structure, and the velocity of glass substrate is higher than that of ZnO film, thus it is expected to have a Sezawa resonance [[22](#_ENREF_19)]. The Rayleigh phase velocities (*vp*) of the fabricated SAW devices (*vp*= *λf*) are 2478, 2580, and 2720 m·s-1, respectively, increasing with the increased wavelength from 16 to 32 μm, as shown in Table 2. When the wavelength is increased, more energy is dispersed in the glass substrate, leading to a higher acoustic velocity of the layered structure, consistent with the observation from other layered structure SAW devices[[23](#_ENREF_20)]. The Sezawa phase velocity also increases from 5120 to 5600 m·s-1 with the wavelength increased from 16 to 32 μm, showing a similar trend with that of the Rayleigh wave.

The electromechanical coupling coefficient, *K*2, can be evaluated using *K*2 = *π Gm (fi)* /4N *Bs (fi)* [[24](#_ENREF_21)], where *N* is the finger pairs, *Gm (fi)* and *Bs (fi)* the motional conductance and static susceptance of the input port at *fi*, respectively. The *K*2 value of Rayleigh wave decreases from 1.80%, 1.31% to 0.5 %, as the wavelength of the devices is increased from 16 to 32 μm. When the wavelength increases, more energy is dispersed in the non-piezoelectric glass, thus leading to a decrease in the *K*2 value. The *K*2 values of the Sezawa waves also decrease from 0.22%, 0.16% to 0.13 % as the wavelength increases from 16 to 32 μm.

It is well known that SAW devices can be used for sensors, microfluidicsand lab-on-a-chip. If the transparent SAW devices can be proven to be useful for high performance actuators or sensors, this will open the door for widespread applications for inexpensive, transparent and invisible sensors and lab-on-a-chip for healthcare, medical research and environment monitoring. When a liquid droplet is located on the path of the surface acoustic wave, the SAW will interact with liquid and the acoustic energy is coupled into the liquid, inducing acoustic streaming inside the droplet. This has been utilized for pumping, mixing, atomization as shown in Fig. 9(a) [25]. The transparent SAW device developed in this study can also deliver the same functions. Figure 9(b) is a snap shot of the acoustic streaming induced by the Rayleigh wave from a transparent SAW device (device C1), showing a stable streaming with a double vortex pattern similar to those obtained from the visible SAW devices on LiNO3 substrates [[26](#_ENREF_22)]. Figure 9(c) shows the measured streaming velocities at the center of the droplet as a function of RF signal voltage and droplet size. The streaming velocity was found to increase with the increase of RF signal voltage applied to the IDT electrode or with the increase of droplet size, and reaches a value of 2.27 cm s-1 at a signal voltage of 45 V and droplet size of 30 μL. The results clearly indicate that the transparent SAW devices can be used for transparent microfluidic and lab-on-chip applications.

1. **Conclusions**

In summary, we investigated the synthesis and characteristics of AZO thin films on glass substrates deposited by DC magnetron sputtering. Results show that a low deposition pressure and optimal sputtering power will yield a good AZO film with (0002) orientation. The optimal deposition parameters for depositing (0002) oriented AZO films were found to be: gas pressure of 0.3 Pa, sputtering power of 300 W, with substrate temperature of 200 °C. The transparent SAW resonators on glass substrates were fabricated using AZO as the electrodes using conventional UV-light photolithography and lift-off process. Transparent SAW devices with different wavelengths exhibited two wave modes of Rayleigh and Sezawa waves, with frequency up to 154.9 and 320 MHz respectively. As the wavelength increases, the resonant frequency of Rayleigh and Sezawa wave increases, while the phase velocity of both the waves decreases. The transparent SAW devices have demonstrated their ability to induce strong acoustic streaming with a streaming velocity up to 2.27 cm/s. This research demonstrated that transparent SAW devices have a great potential for applications in transparent electronics, microfludics, sensors and microsystems.

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**Table captions**

**Table 1.** Deposition parameters for AZO thin films by DC magnetron sputtering

|  |  |
| --- | --- |
| Sample No. | Deposition Parameters\* |
| Deposition pressure(Pa) | Sputtering power(W) | Target-substrate distance(mm) | Substrate temperature (oC) |
| A1 | 0.3 | 300 | 70 | 200 |
| A2 | 0.5 |
| A3 | 1.0 |
| A4 | 1.5 |
| B1 | 0.3 | 100 |
| B2 | 150 |
| B3 | 200 |
| B4 | 300 |
| B5 | 400 |

\* Other deposition parameters are: AZO target (ZnO 98 wt.%: Al2O3 2 wt.%,) with a diameter of 50 mm

**Table 2**. The effect of wavelength on the characteristics of transparent SAW devices

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| sample no. | λ (μm) | *f*1 (MHz) | *v*p0 (ms-1) | *f*2 (MHz) | *v*p1 (ms-1) |
| C1 | 16 | 154.9 | 2478 | 320 | 5120 |
| C2 | 20 | 129 | 2580 | 272 | 5440 |
| C3 | 32 | 85 | 2720 | 175 | 5600 |

**List of figure captions**

Fig. 1 The effects of deposition pressure on (a) the intensity of XRD; and (b) the FWHM of XRD and grain size for (0002) AZO thin films.

Fig. 2 The effect of deposition pressure on resistivity, Hall mobility and carrier concentration of the AZO thin films.

Fig. 3 Optical transmittance of AZO thin films deposited at different deposition pressures.

Fig. 4 SEM images of AZO thin films deposited at different powers: (a) 100 W, (b) 150 W, (c) 200 W, (d) 300 W, (e) 400 W; (f) The deposition rate of AZO as a function of deposition power.

Fig. 5 The effect of deposition power on the FWHM of the XRD pattern and grain size of AZO films.

Fig.6 A three-dimensional schematic of the transparent SAW device; (b) the SEM picture of the ZnO/AZO film on the transparent glass; (c) a photograph of a glass wafer with fabricated transparent SAW devices; (d) and a microscope image of a SAW device with a 20 pairs of AZO IDT fingers.

Fig.7 Transmission spectrum of the transparent AZO/ZnO/glass SAW devices as a function of wavelength, λ, (a) λ=16 μm; (b) 20 μm; (c) 32 μm. Two well resonant modes with high amplitude were obtained from all devices with different wavelengths.

Fig. 8 Geometry of a periodic cell in the simulation; (b) simulated Impedance of the transparent AZO/ZnO/Glass SAW device and (c) particle vibration and surface deformation of the two modes.

Fig. 9 (a) Schematic drawing of acoustic streaming inside a liquid droplet, (b) snapshot of acoustic streaming induced by transparent SAW device, showing double vortex patterns. The yellow arrow indicates the SAW direction entering the liquid, (c) and the effect of RF signal voltage and droplet size on the streaming velocity, showing rapid increase in streaming velocity with signal voltage and droplet size.