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# Photoluminescence of ZnO thin films deposited at various substrate temperatures

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#### ABSTRACT

This study investigated surface acoustic wave devices with an Al/ZnO/Si structure for use in ultraviolet sensors. ZnO thin films were fabricated using a reactive radio frequency magnetron sputtering system. The substrate temperature of ZnO thin films can be varied to obtain highly crystalline properties. The surface morphologies and c-axis preferred orientation of the ZnO thin films were determined using scanning electron microscopy and X-ray diffraction. In addition, bright-field images of ZnO crystallization were investigated using a transmission electron microscope. From photoluminescence analysis, four peaks were obtained at 377.8, 384.9, 391.4, and 403.4 nm. Interdigital transducers of an aluminum electrode were fabricated on the ZnO/Si structure by using a direct current sputtering system and photolithography, combined with the lift-off method, thereby obtaining a surface acoustic wave device. Finally, frequency responses were measured using a network analyzer, and an illuminating test was adopted for the ultraviolet sensor, using a wavelength of 355 nm from a light-emitting diode. The sensitivities of the ultraviolet sensor were also discussed.

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#### 1. Introduction

Surface acoustic wave (SAW) devices are small, lightweight, lowcost, strong, and have high sensitivity; they have therefore been studied for use in sensors [1–3]. SAW devices are based on an interdigital transducer (IDT), piezoelectric thin films, or a piezoelectric substrate. In this study, SAW devices with an IDT, piezoelectric thin films, and a piezoelectric substrate were used because such devices generate the Sezawa mode at a higher velocity and a higher sensitivity than the Rayleigh mode [4,5]. Piezoelectric materials such as lead zirconate titanate, aluminum nitride, and zinc oxide (ZnO) are used in acoustic devices for various applications [6–10]. ZnO is a II–VI semiconductor with a wurtzite hexagonal crystal structure. Because it is not a ferroelectric material, ZnO thin films with a c-axis orientation are the only ZnO materials to exhibit a piezoelectric effect. In this study, ZnO piezoelectric thin films were used in SAW devices because of their high thermal stability, large band gap, high electrical resistivity, and high electromechanical coupling factor, among other favorable properties [11,12].

Ying *et al.* and Hong *et al.* fabricated ZnO nanowires for ultraviolet (UV) sensors and found that their resistivity changed from 1.7 to 0.2  $\Omega$ -cm on irradiation by UV illumination [13]. Because of this change in resistivity, ZnO thin films are adopted in SAW UV sensors [14–16]. In addition, the phase measurement method is adopted because it can be used to observe the relationship between the UV sensor response and

\* Corresponding author. *E-mail address:* dlcheng@stu.edu.tw (D.-L. Cheng). duration of UV light irradiation [17]. However, the LiNbO<sub>3</sub> substrate adopted in previous research is expensive and prevents cost reduction. A ZnO-based SAW device using silicon (Si) as a low-priced substrate is proposed for use in UV sensors.

#### 2. Experimental details

In this study, SAW devices for UV sensors were constructed with an IDT electrode and a ZnO/Si structure. In each device, the substrate was a 4-in p-type (100) Si wafer, which was diced into  $2 \times 2$  cm pieces. After the substrates were cleaned in a standard process, ZnO thin films were deposited on them by using a reactive radio frequency (RF) magnetron sputtering system with a Zn (99.995%) target with a 2-in diameter in a mixed atmosphere of argon and oxygen. Table 1 presents the deposition parameters that yielded ZnO thin films with excellent piezoelectric properties and a uniform crystal size. The crystalline characteristics of ZnO thin films were measured through X-ray diffraction, using a Siemens D8 with CuK $\alpha$  radiation. The diffraction angles of the ZnO thin films were obtained by scanning between 20° to 60° at a speed of 0.05°/s. The surface morphologies, cross-sectional morphology, and crystallization of ZnO thin films were observed using a field emission scanning electron microscope and a transmission electron microscope (TEM) at operating voltages of 10 kV and 150 kV, respectively.

The diverse luminescent characteristics of the ZnO thin films were investigated according to photoluminescence (PL) measurements obtained using an He-Cd laser with a wavelength of 325 nm.







Table 1Please check the presentation of Tables 1 and 2 and amend if necessary
Deposition parameters of ZnO thin films and Al IDT electrodes.

Sputter system	Reactive RF	DC
Target	Zn (99.995%)	Al (99.995%)
Base pressure (mPa)	<0.4	< 0.13
Power (W)	120	100
Sputtering pressure (Pa)	6.67	0.53
Substrate temperature (°C)	R. T. ~350	R. T.
$O_2/O_2 + Ar ratio (\%)$	75	0
Total gas flow (sccm)	7.5	10

Al thin films were deposited on the ZnO/Si substrate as IDT electrodes by using a direct current sputtering system, and photolithography and lift-off methods were then employed. Table 1 presents the deposition parameters of the Al thin films. Table 2 presents the design parameters of the IDT electrodes for SAW devices.

Finally, the frequency responses of were measured using a network analyzer (Agilent N5230A) and power supply (GW Instek PST-3202). A UV lamp and measurement apparatus were set up in an airtight box, as displayed in Fig. 1(a). Fig. 1(b) presents the UV light illumination and measurement details. The emitted wavelength of the UV lamp (Sensor Electronic Technology) was 355 nm; the working voltage was fixed at 4 V; the distance between the light source and SAW devices was 10 mm, and the UV intensity was 2000  $\mu$ m/cm<sup>2</sup>. The phase difference of the SAW device signals was measured using a network analyzer by feed specify the frequency of the RF signals.

#### 3. Results and discussion

The substrate temperature that optimized the characteristics of the ZnO thin films was obtained to achieve highly c-axis-oriented ZnO thin films.

#### 3.1. Influence of substrate temperature on ZnO crystal properties

The substrate temperature was varied to determine the optimal deposition parameters for ZnO thin films, which are provided in Table 1. The substrate temperature was varied from room temperature (RT) to 350 °C to ensure a strong c-axis orientation. Fig. 2 presents a plot of the various substrate temperatures at a sputtering pressure of 6.67 Pa. A substrate temperature of 300 °C yielded a ZnO (002) diffraction peak, revealing the strong c-axis (002) orientation of the piezoelectric thin films and the consequently high electromechanical coupling coefficient.

Fig. 3 presents the variations in the surface structure and crosssectional morphologies with an increasing substrate temperature. The density of grain boundaries decreased and the grain size increased as the substrate temperature increased. However, the surface morphologies exhibited melting and recrystallization on grain overgrowth at a substrate temperature of 350 °C.

The ZnO thin films that were deposited at RT, 200 °C, and 300 °C exhibited uniform cobblestone crystalline structures and well-textured columnar cross-sectional structures. Therefore, a sputtering pressure of 6.67 Pa, a substrate temperature of 300 °C, an RF power of 120 W, and a gas ratio of 75% were used to optimize the characteristics of the

#### Table 2

The designed parameters of IDT electrodes for the SAW device.

Device type	Delay line
Single/two ports	Single
Input IDT electrode (pairs)	32 30
Output IDT electrode (pairs)	30
Length of IDT electrode (µm)	2816
The overlapped length of IDT electrode (µm)	2752
IDT electrode of Al layer thickness (µm)	0.1



Fig. 1. (a) Airtight box system (b) UV light illumination and measurement details.

ZnO thin films. A ZnO thin film with highly c-axis (002)-oriented crystallization, a well-textured columnar structure, and a thickness of approximately 1.8 µm was obtained.

#### 3.2. Optical characteristics of ZnO thin films

The optical characteristics of ZnO thin films were investigated on the basis of PL spectra. Fig. 4 displays PL spectra of the ZnO thin films formed at various substrate temperatures. The height of the PL peaks increased with the substrate temperature of the ZnO thin film. Depending on the deposition parameters, the two most common defects in ZnO thin films are oxygen and zinc vacancies [18–20]. PL indicated that the emission behavior of the ZnO thin films fabricated at 300 °C was strong at a



Fig. 2. XRD pattern of ZnO thin films deposited with various substrate temperatures.

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