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Annealing effect on the generation of dual mode acoustic waves in inclined ZnO films

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ABSTRACT

ZnO films with different inclined angles on steel substrates were sputter-deposited by changing the substrate tilt angle during deposition and then used to fabricate ZnO film ultrasonic transducers. The ultrasonic performance of those devices was characterized using a standard pulse-echo method. A dual mode wave with both longitudinal and shear wave components was detected from the ZnO device at 0° inclined angle. At a columnar inclined angle of 31°, longitudinal wave excitation was suppressed with a nearly pure shear wave detected. Post annealing of the ZnO film improved the crystallinity and decreased the film stress. The dispersion of the received echoes was observed when the grain sizes of ZnO films were increased after annealing. The frequency components of the waveforms were analyzed and identified using a short time Fourier transform. Post-annealing of the ZnO films changed the primary frequency and enhanced the propagation of the relative high-frequency acoustic wave.

1. Introduction

Recently ZnO films have been widely studied for high performance acoustic wave based microsensors and microfluidics for lab-on-chip and environmental devices. Benefits include low cost, fast response, reduced reagent requirement and increased precision [1–3]. ZnO films normally grow in a hexagonal or wurtzite type crystalline structure and the (0002) plane has the lowest surface free energy [4]. Therefore, in the absence of epitaxy between film and substrate, and without any external ion or plasma source, ZnO films grow in the (0002) orientation on many different substrates. ZnO acoustic wave devices with a (0002) film texture can be applied for sensing in air, gaseous and dry environments [1]. However, biosensors are often needed to detect chemical reactions in a liquid environment. If liquid is present on the sensing surface, excessive damping of the propagating wave occurs when the longitudinal mode (L-mode) wave couples into the liquid [5]. A common solution is to use an in-plane shear horizontal (SH) mode wave, thus dramatically reducing wave coupling into a liquid medium [6], but this normally requires deposition of the ZnO films with special orientations to generate the SH mode wave [7,8]. On the other hand, deposition of *c*-axis inclined films allows both longitudinal and shear wave modes (S-mode) to be generated on a single device with different frequencies which could be individually controlled for microfluidic or sensing purposes [1,9–11].

Common methods used to deposit inclined ZnO films by magnetron sputtering include varying the substrate-tilting angle or the angle between the substrate and target [12,13]. Theoretical analysis has shown that pure thickness L-mode of a ZnO based acoustic devices with maximum coupling coefficient occurs at inclined angles of 0° and 65.4°, and maximum amplitude S-mode occurs at 41° [14]. A systematic experimental study on changes of wave modes as a function of film inclined angles (especially above 30°) for sputtered ZnO films has seldom been reported, and the annealing effect on the dual mode wave generation and propagation is not discussed.

In this paper, we report that by changing substrate-tilt angle, ZnO films with different inclined crystal angles were deposited on ferritic carbon steel. Film ultrasonic transducers based on those films were fabricated and tested using an ultrasonic pulse-echo method. Annealing effect on the ultrasonic properties of the ZnO film transducers was investigated. Ultrasonic frequency analysis was performed using short time Fourier transformation (STFT) to identify the propagation modes and the multiple frequencies component were discussed.

2. Experimental

ZnO films were deposited on ferritic carbon steel using DC magnetron sputtering from a rectangular Zn target (100 mm ×

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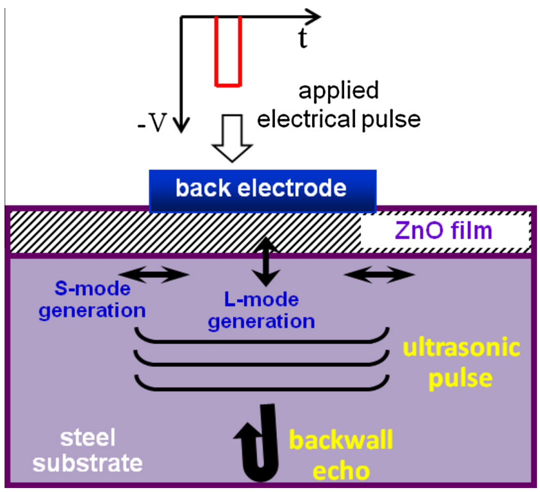

Fig. 1. Schematic of pulse echo experiment using the ZnO film deposited on ferritic steel plate, showing generation and propagation of longitudinal wave (L-mode) and shear wave (S-mode). Figure indicates generation of S-mode at edges of electrode by *c*-axis films with 0° inclined angle.

300 mm) at a power of 400 W, an Ar/O_2 flow ratio of 90/45 sccm (standard cubic centimeter per minute) and a deposition temperature of 150 °C. Gas pressure during deposition was 4.8 mTorr. Substrates were tilted at different angles using a specially designed multi-angle plate holder, in order to obtain ZnO films with different inclined angles in one deposition whilst maintaining a similar distance from the target.

The morphology and crystallinity of the films were characterized using scanning electron microscopy (SEM, Hitachi S4100) and X-ray diffraction (XRD, Siemens D5000 Cu Kα, 40 kV/30 mA), respectively. The average grain size was estimated from peak broadening using the Debye–Scherrer equation based on the XRD analysis with correction for instrumental broadening. Film stress was estimated using Eq. (1) based on XRD peak shift [15]:

$$\sigma_f = \frac{E_f}{2\nu_f}\frac{\Delta 2\theta}{2\tan\theta_o} \quad (1)$$

where E_f and ν_f are the Young's modulus and Poisson ratio of the ZnO films (124 GPa and 0.3, respectively), θ_o is the Bragg angle of stress free ZnO, θ is the diffraction angle, and $\Delta 2\theta = 2(\theta - \theta_o)$.

In order to evaluate the piezoelectric properties of the ZnO films, pulse-echo experiments were performed using an ultrasonic pulser/receiver (JSR Ultrasonics DPR300, 475V, USA) as illustrated in Fig. 1. The ZnO film was deposited on ferritic steel plates (area of $25 \times 25\ mm^2$ and thickness of 2.9 mm) and used to fabricate the film based transducer with a top electrode of silver paste (diameter of 2 mm). A negative spike pulse with amplitude of 100 V and duration of 0.2 μs was generated and applied to the ZnO film transducer. The film was excited by the pulse, and echo signals were produced and reflected by the back wall of the steel substrate when the ultrasonic wave propagated in the steel substrate. The receiver was set to a gain of 50 dB and a low frequency pass filter at 50 MHz. The high frequency pass filter was switched off. The echo signals were received from the ZnO film transducer. The electrical signal from the pulse/receiver was recorded using a digital oscilloscope (Agilent 54641A, Agilent Technologies UK Ltd., UK). All the equipments of the measurement system were connected using 50 Ω coaxial cables. Some of the films were annealed at 400 °C for 1 h in a furnace in air, and then were characterized using XRD. The transducers with post-annealing ZnO films were fabricated and measured using the above pulser/receiver system. In order to analyze the wave frequency, a STFT of the acoustic wave were performed for all the echo signals.

3. Results and discussions

From SEM observation, the ZnO films deposited with substrate tilt angles of 0–60° show compact columnar structures as shown in Fig. 2a–e. The film surface shows equi-axial crystals for these films in Fig. 2f–h. This is because the ZnO crystals typically grow in a hexagonal wurzite structure and form long rods along the *c*-axis resulting in columnar grain structure. With tilting of the substrate during deposition, the ZnO columnar structure shows

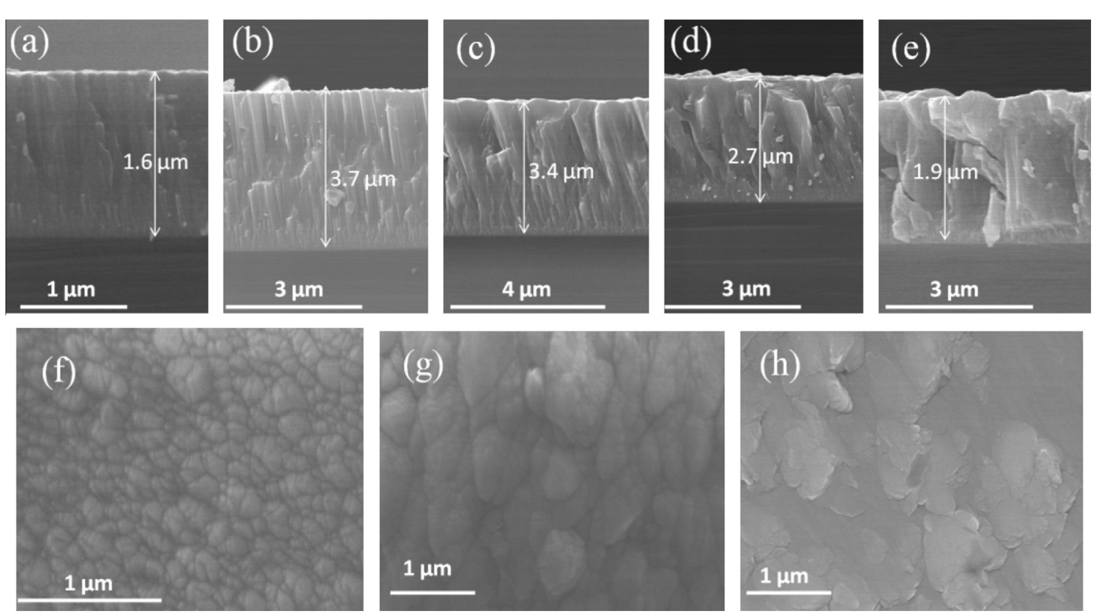

Fig. 2. (a–e) Cross-sectional SEM images of ZnO films deposited at substrate tilt angle from 0° to 60°, and surface morphologies (f–h) of ZnO films with substrate tilt angles of 0°, 30° and 60°, respectively.