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Fabrication of Ultrasonic Transducers Using Epitaxial $Pb(Zr,Ti)O_3$ Thin Films on Epitaxial γ-Al_2O_3/Si Substrates for Smart Sensors

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Ultrasonic transducers using epitaxial $Pb(Zr,Ti)O_3$ (PZT) thin films on epitaxial γ-Al_2O_3/Si substrates were successfully fabricated for the first time. The characteristics of PZT thin films on the γ-Al_2O_3/Si substrates were investigated. 240-nm-thick PZT films with various compositions were formed by the conventional sol-gel method. All PZT films were epitaxially grown on the substrates and exhibited ferroelectric and piezoelectric characteristics. Ultrasonic transducers were fabricated on the epitaxial γ-Al_2O_3(001)/Si(001) substrates with transducer element of 1 mm square. The transmission and reception characteristics of the ultrasonic transducers were observed in water by a hydorophone. Fabricated transducers can transmit and receive an ultrasonic wave with frequency of 2.5 MHz at distances of 15 and 20 mm. From these results, ultrasonic transducers with the epitaxial PZT/Pt/γ-Al_2O_3/Si structure can be applied to Si monolithic ultrasonic smart sensors.

1 Introduction

Ultrasonic sensors are widely used for various sensing applications, such as, medical diagnostics systems, object detection for fish finding, automobile control, robot eyes and security systems. These applications require high resolution with high-scanning frequency, microscale probe size, and two-dimensional arrays for high directivity and easy observation of three-dimensional objects. $Pb(Zr,Ti)O_3$ (PZT) ceramics are used for ultrasonic transducers due to their superior piezoelectric characteristics and electrome-

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chanical properties. On the other hand, it is very hard to fabricate 2D micro-PZT ceramics array for ultrasonic sensors. Some types of ultrasonic sensors on silicon substrates have been developed using the principle of the piezoresistive effect of a silicon diaphragm made by a conventional large-scale integration (LSI) process;[1] a condenser microphone-type sensor using a micromachined silicon diaphragm[2] and piezoelectric sensors.[3,4] However, few studies were reported in the integration of piezoelectric ultrasonic sensors with signal processing circuits.

Meanwhile, we have studied epitaxial γ-Al_2O_3 films on silicon substrates using chemical vapor deposition[5] and molecular beam epitaxy.[6] Epitaxial γ-Al_2O_3 films on Si substrates exhibit desirable features such as chemical and physical stability and good interface characteristics with Si.[5] γ-Al_2O_3 films have wide application, such as insulation for silicon-integrated devices[7,8] and as a buffer layer for heteroepitaxial growth.[9–12] Using a crystalline insulator on Si substrates is useful for controlling the orientation of piezoelectric films and the integration of signal processing circuits.

In this paper, we propose epitaxial γ-Al_2O_3 films on Si substrates to control the orientation of Pt and PZT films. We investigate the ferroelectric properties of epitaxial PZT films with various compositions on the substrates and fabricate ultrasonic transducers using the epitaxial PZT/Pt/γ-Al_2O_3/Si structure for the first time.

2 Experimental Procedure

2.1 *Preparation of PZT thin films*

The fabrication of epitaxial γ-Al_2O_3 films on Si substrates was carried out using the CVD method.[5] γ-Al_2O_3(001) and γ-Al_2O_3(111) films 60 nm thick were epitaxially grown on Si(001) and Si(111) substrates, respectively. The characterization of the γ-Al_2O_3 films has been reported elsewhere.[5,6] Epitaxial Pt thin films were prepared by rf sputtering using a Pt (99.95%) target and Ar gas.[12] (001) and (111) γ-Al_2O_3/Si substrates were coated with (001) and (111) epitaxial Pt films 100 nm thick, respectively. Polycrystalline Pt films with the same thickness were deposited at room temperature on the γ-Al_2O_3(001)/Si(001) substrates. The 240-nm-thick PZT films were prepared by the sol-gel method. The PZT precursor solution with various compositions (Zr/Ti ratios of 20/80, 40/60, 52/48, 60/40 and 80/20) supplied by Mitsubishi Material Corp. were used. Three sol layers were spin-coated; each layer was dried at 150°C for 2 min, subsequently prebaked in air at 450°C for 15 min and crystallized at 650°C for 30 min in O_2 ambient.

The crystal structure and the orientation of the films were characterized using X-ray diffractometer (XRD). The ferroelectric properties and piezoelectric butterfly curves of the PZT films were measured using a FCE-1 ferroelectric evaluation system (Toyo) with atomic force microscopy by fabricating 100 mm square Pt top electrodes on the PZT films.

2.2 *Fabrication of ultrasonic transducers*

Epitaxial PZT/Pt/ γ-Al_2O_3(001)/Si(001) structures were fabricated as described above. The Zr/Ti ratio of the PZT films was 52/48 (morphotropic phase boundary composition). A hundred-nm-thick Au films were deposited on the PZT film for top electrode by vacuum evaporation. The top Au electrode and PZT films were formed with a size of 1 mm square using conventional photolithography and an inductive-coupled plasma reactive ion etching machine. After etching, the chips were diced to a few mm square and mounted on a ceramic die. Au wire and Ag paste were attached to the top Au electrode and bottom Pt electrode to connect the electrodes of the chips and the terminal pad of ceramic die. A schematic illustration of a typical sample structure is shown in Fig. 1. The transducers had no structure for backing and matching layers. The transmission and reception characteristics of the PZT/Pt/γ-Al_2O_3/Si ultrasonic transducers were investigated using the impulse response in the 2.5 MHz frequency band by the hydrophone method. The experimental setup is shown in Fig. 2.

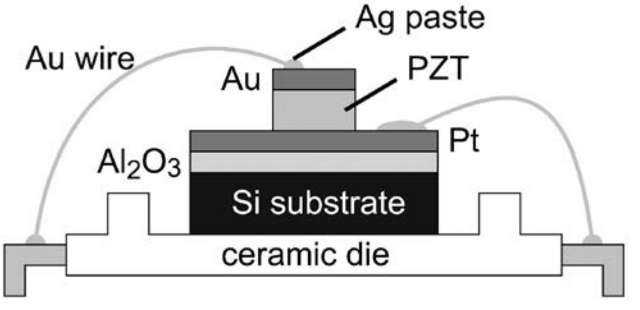


Fig. 1. Schematic illustration of ultrasonic transducer using epitaxial PZT/Pt/γ-Al_2O_3/Si structure.

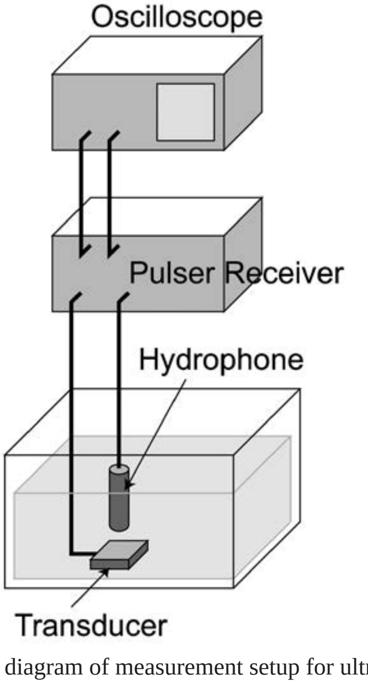


Fig. 2. Schematic diagram of measurement setup for ultrasonic transducer.

3 Results and Discussion

3.1 *PZT films*

Figure 3 shows the XRD patterns from the PZT films with a Zr/Ti ratio of 52/48 on various substrates. These PZT films have a perovskite crystalline structure and no pyrochlore phase is observed. The XRD pattern of sample (a) has a strong (00l) peak and the film is (001)-oriented. The XRD pattern of sample (b) has a strong (111) peak and a weak (110) peak. This implies that the film is (111)-oriented. This shows that highly oriented PZT films can be fabricated on epitaxial Pt/γ-Al_2O_3/Si substrates using the sol-gel method. Sample (c) showed many perovskite PZT peaks and that indicates the formation of polycrystalline PZT films. (001)-oriented, (111)-oriented and polycrystalline PZT films were also obtained using other compositions of PZT sol-gel precursors (not shown).

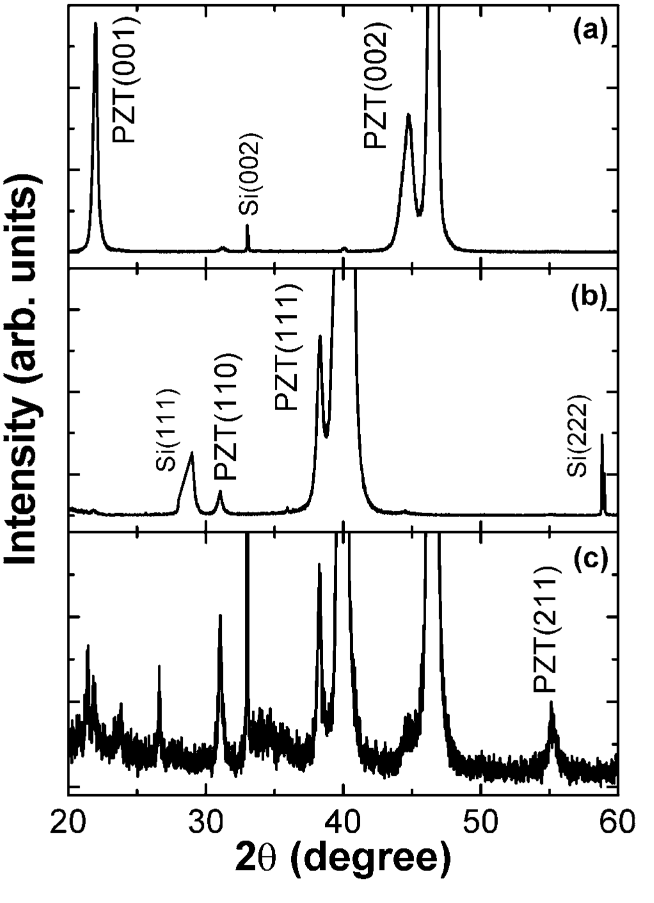


Fig. 3. XRD patterns of PZT (Zr/Ti = 52/48) films on (a) epitaxial Pt(001)/ γ-Al_2O_3(001)/Si(001) substrates, (b) epitaxial Pt(111)/ γ-Al_2O_3(111)/Si(111) substrates and (c) polycrystalline Pt/ γ-Al_2O_3(001)/Si(001) substrates.

Figure 4 shows *P-E* hysteresis loops for PZT films of various orientations with a Zr/Ti ratio of 40/60. Saturated hysteresis loops were observed for all films. Values of remnant polarization (2*Pr*) and coercive field (*Ec*) are 50–60 μC/cm^2 and 65–75 kV/cm, respectively, at a maximum applied voltage of 10 V. Initial polarization values of epitaxial PZT films were higher than that of polycrystalline PZT film. Furthermore, polycrystalline PZT films had large *Ec* values and poor *P-E* loop shapes compared with those of epitaxial PZT films. This suggests that the polarization directions of the as-deposited epitaxial PZT films were oriented. Values of 2*Pr* and *Ec* from various compositions and orientations of PZT films are shown in Fig. 5. Epitaxial PZT(001), PZT(111) and polycrystalline PZT films show the same tendency of 2*Pr* values decreasing with increasing Zr content. These results show the same tendency as previously reported results.[13]

Figure 6 shows displacement values from butterfly curves of various orientations and compositions of PZT films on Pt/γ-Al_2O_3/Si substrates. The maximum displacement values of epitaxial PZT films were observed for a Zr/Ti ratio of 52/48 for the PZT(111) film and that of 40/60 for the PZT(001) film. These values were about 0.8 nm and equivalent to 0.3% of the film thickness. This is a good value as compared with the PZT ceramics. On the other hand, displacement values of polycrystalline PZT films were less than those of epitaxial PZT films. Other PZT films with various compositions showed the same tendency as shown in Fig. 6. These epitaxial PZT films can be applied to Si integrated ultrasonic transducers.

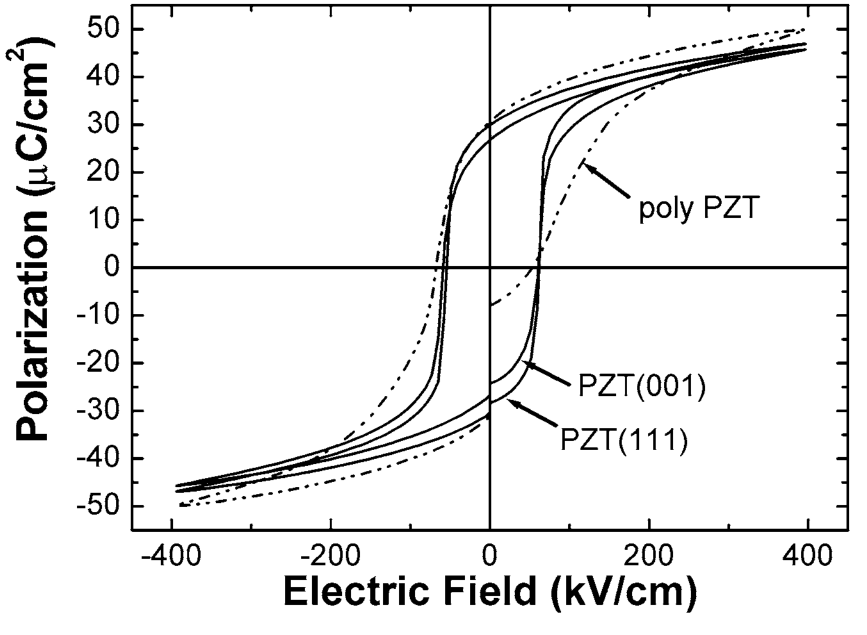


Fig. 4. *P-E* hysteresis loops of PZT (Zr/Ti ratio of 40/60) films with different orientations.

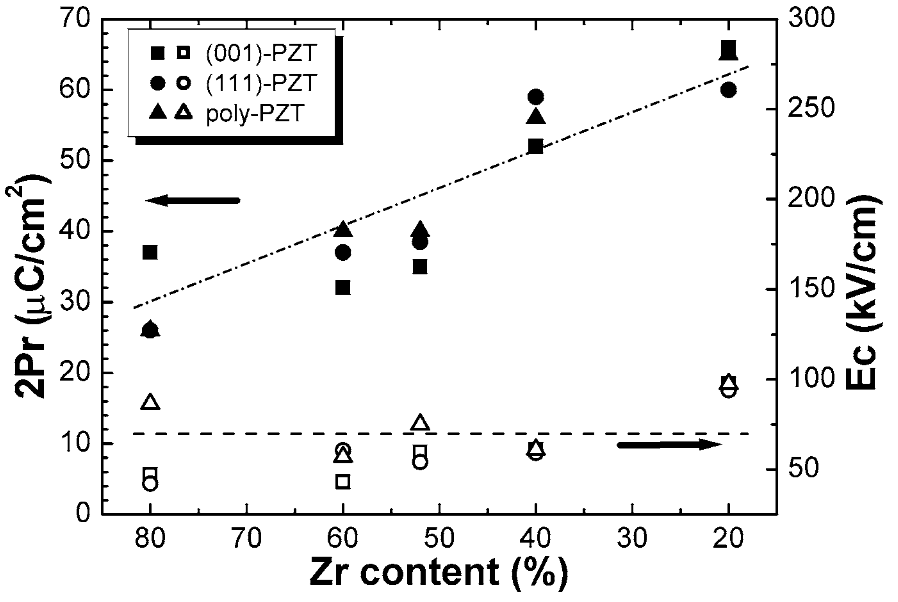


Fig. 5. Zr content dependence of remnant polarization (2*Pr*) and coercive field (*Ec*) for PZT films of various orientations.

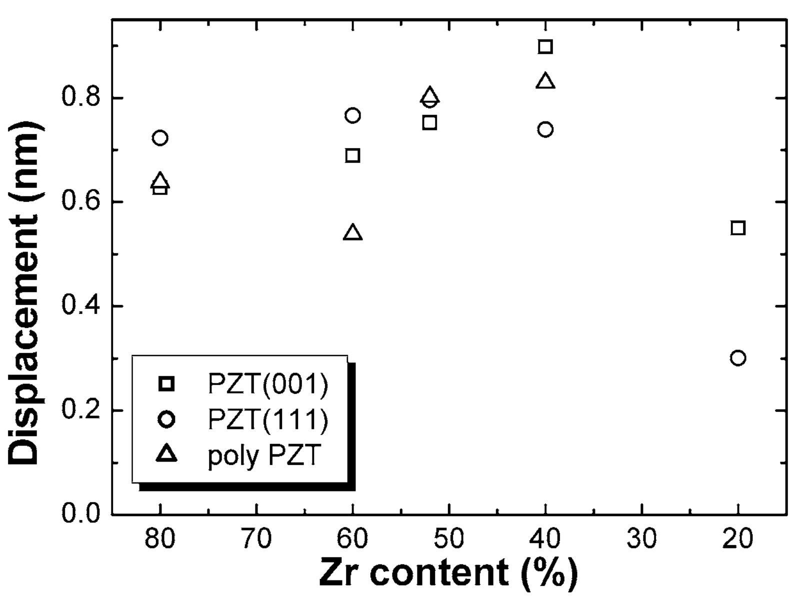
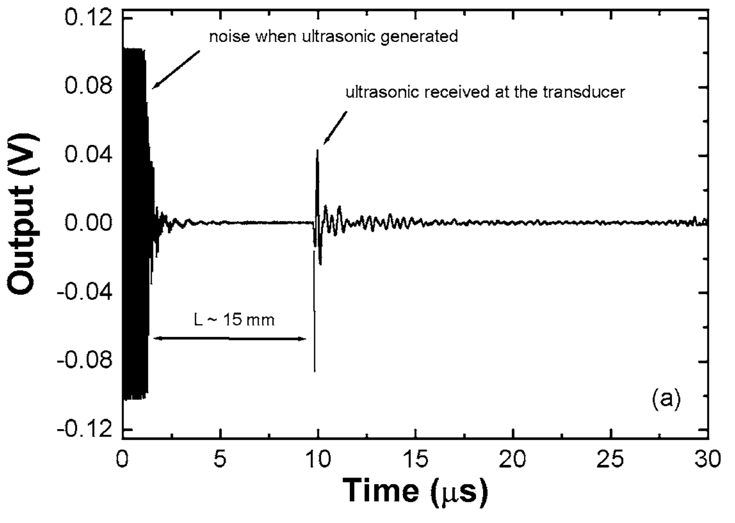


Fig. 6. Displacement values from butterfly curves of various orientations and compositions of PZT films on Pt/ γ-Al_2O_3/Si substrates.

3.2 *Transducer*

A hydrophone or fabricated transducers were driven by a signal from a pulser receiver system. The generated ultrasonic propagated in the water. The ultrasonic was detected by the hydrophone for the transmission test and the fabricated transducers for the reception test. The detected wave signal was amplified by a pulser receiver at 30 dB, and this signal was displayed and recorded on an oscilloscope. Figure 7 shows ultrasonic signal pulses detected by the fabricated transducers that radiated from the hydrophones at distance of

(a)



(b)

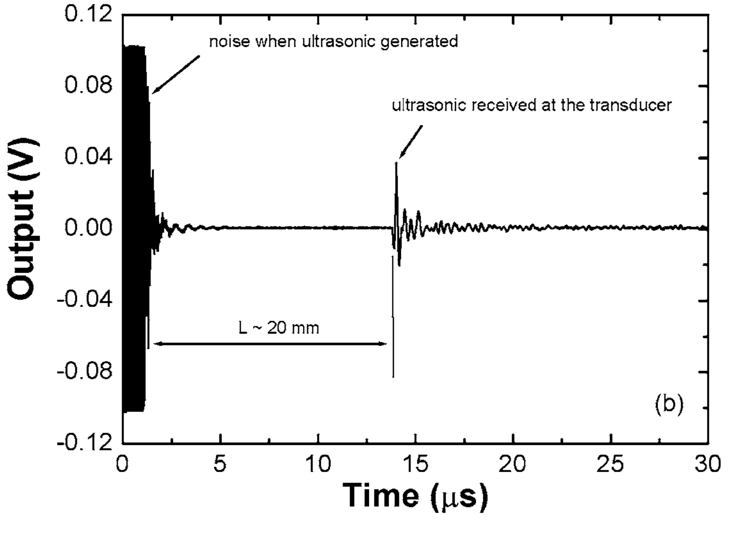


Fig. 7. Ultrasonic single pulse detected by fabricated transducers that radiated from hydrophone at distance of approximately (a) 15 nm and (b) 20 nm.

approximately 15 and 20 mm. The propagation times were approximately 10 and 14 μs at distances of 15 and 20 mm, respectively. This result indicates that the transducers directly received the ultrasonic from the hydrophone and show the possibility of distance measurement. The ultrasonic radiating from the fabricated transducers was investigated by the hydrophone at a distance of approximately 20 mm. The detected ultrasonic waveform is shown in Fig. 8. The propagation time was approximately 14 μs. We succeeded in the transmission and reception of ultrasonic waves using epitaxial PZT thin-film transducers on epitaxial γ-Al_2O_3/Si substrates for the first time.

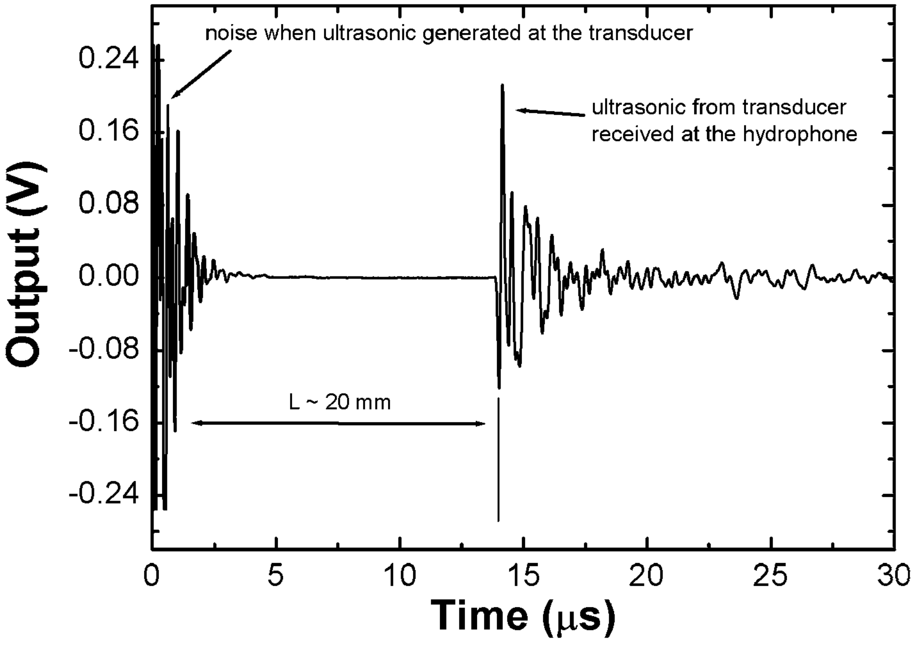


Fig. 8. Ultrasonic single pulse detected by hydrophone that radiated from fabricated transducer at distance of approximately 20 mm.

4. Conclusion

Ultrasonic transducers using epitaxial PZT thin films on epitaxial γ-Al_2O_3/Si substrates were successfully fabricated for the first time. The characteristics of PZT thin films on the γ-Al_2O_3/Si substrates were investigated. 240-nm-thick PZT films with various compositions were formed by the conventional sol-gel method. All PZT films were epitaxially grown on the substrates and exhibited ferroelectric and piezoelectric characteristics. Epitaxial PZT films on the γ-Al_2O_3/Si substrates exhibited larger spontaneous polarization than the polycrystalline PZT thin films. Ultrasonic transducers were fabricated on the epitaxial γ-Al_2O_3(001)/Si(001) substrates using $Pb(Zr_{0.52},Ti_{0.48})O_3$ thin film with a transducer element of 1 mm square. The transmission and reception characteristics of the ultrasonic transducers were observed in water by a hydrophone. Fabricated transducers can transmit and receive an ultrasonic wave with frequency of 2.5 MHz at distances of 15 and 20 mm. From these results, ultrasonic transducers with the epitaxial PZT/Pt/γ-Al_2O_3/Si structure can be applied to Si monolithic ultrasonic smart sensors.

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