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To cite this article: M. D. Nguyen *et al* 2017 *J. Phys.: Conf. Ser.* **922** 012022

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Wafer-scale growth of highly textured piezoelectric thin films by pulsed laser deposition for micro-scale sensors and actuators

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Abstract. Piezoelectric lead-zirconate-titanate (PZT) thin films were deposited on 4-inch (111)Pt/Ti/SiO$_2$/Si(001) wafers using large-area pulsed laser deposition (PLD). This study was focused on the homogeneity in film thickness, microstructure, ferroelectric and piezoelectric properties of PZT thin films. The results indicated that the highly textured (001)-oriented PZT thin films with wafer-scale thickness homogeneity (990 nm ± 0.8%) were obtained. The films were fabricated into piezoelectric cantilevers through a MEMS microfabrication process. The measured longitudinal piezoelectric coefficient (d_{33f} = 210 pm/V ± 1.6%) and piezoelectric transverse coefficient (e_{31f} = -18.8 C/m^2 ± 2.8%) were high and homogeneity across wafers. The high piezoelectric properties on Si wafers will extend industrial application of PZT thin films and further development of piezoMEMS.

1. Introduction

Pulsed laser deposition (PLD) differs from other physical vapour deposition techniques because of the very high kinetic energy of the ablated particles arriving at the substrate and the high deposition rate during each laser pulse. Already since 1965, Smith and Turner utilized a pulsed ruby laser to deposit the first vacuum deposited thin films [1], the PLD technique is only widely known when Dijkkamp *et al*. [2] were able to laser-deposit a thin film of YBa$_2$Cu$_3$O$_{7-\delta}$, a high-temperature superconductor material, which was of superior quality to that of films deposited with alternative techniques. Nowadays, PLD has become the deposition method to fabricate and study, in a research laboratory environment, epitaxial and single crystalline thin films of complex oxides, such as superconducting high-T$_c$ cuprates, metaloxides, ferroelectric, ferromagnetic, and dielectric oxides. The success of PLD is due to the fact that it is conceptually as well as operationally a relatively simple technique. The main advantage of PLD is the possibility to transfer stoichiometrically multicomponent target material, especially containing volatile components such as lead, to the layer. Moreover, PLD is a powerful method which allows a thicker film to be fabricated in a short time due to the high deposition rate and the possibility to incorporate the process directly into a Si-production line [3, 4], for example in MEMS. Despite the fact that PLD technique has provided an excellent tool for producing high-quality multicomponent films, it has not yet fully emerged as a reproducible process for commercial thin-film applications. This is mainly because the deposited films have only a small area of structural and

thickness uniformity (samples of 10×10 mm^2 or smaller). Especially for the fabrication of MEMS, it is necessary to control the growth of these layers on silicon wafer-scales and to develop appropriate fabrication processes to realize micromachined structures [5, 6].

Microelectromechanical systems (MEMS) actuators driven by lead zirconate titanate at the morphotropic phase boundary, $Pb(Zr_{0.52}Ti_{0.48})O_3$ (abbreviated as PZT), have received wide attention recently because they could potentially outperform other MEMS actuators due to the remarkable high ferroelectric polarization and piezoelectric coefficients of PZT [7-11]. For the integration of PZT-based materials into MEMS these should be prepared in thin film form. Therefore the properties of PZT thin films are critical to the quality and reliability of MEMS devices. For most MEMS device applications, such as vibration energy harvesters, micropumps, microcantilever-based mass sensors and ultrasonic transducers for medical and sonar applications, the transverse piezoelectric coefficient (e_{31} or d_{31}) is the most important factor to be considered. For specific applications, however, such as a nano-control system for the control of optical cavities, a high longitudinal piezoelectric coefficient (d_{33}) is required to obtain a large piezoelectric, piston-like deformation in the PZT film actuators.

The subject of the present study is the deposition of $Pb(Zr_{0.52}Ti_{0.48})O_3$ (PZT) thin films on 4-inch platinized silicon wafers (Pt/Ti/SiO$_2$/Si or short Pt/Si) using a large-area PLD tool developed by Solmates B.V. (a spin-off of the University of Twente–The Netherlands, was founded in 2007 and delivered the first large-area production tool for deposition of complex-oxide thin films on 4-8 inch wafers in 2013) [12]. The thickness homogeneity and microstructure uniformity of 1-µm-thick PZT thin films across wafers have been investigated. The piezoelectric MEMS devices, such as micro-cantilevers and micro-diaphragms were also fabricated. In addition, the high and uniformity of ferroelectric and piezoelectric properties of devices in the entire wafers indicated that the high-quality of PLD-based PZT films on Si wafer-scales will enhance the practical applicability of the piezoelectric MEMS devices.

2. Experimental procedure

Figure 1 shows a photograph of a Solmates's large-area PLD system in MESA+ NanoLab's cleanroom (University of Twente, The Netherlands). This system can handle wafers up to 200 mm in diameter and is specifically designed for PZT deposition, as well as the other perovskites and oxide materials, for MEMS applications. It includes wafer heating up to 800 °C, particle reduction filter, multi-target manipulator, fully automated with SEMI compatible fab interface software, integrated laser with homogenizer and beam attenuator. The fast growth rate of 3.4 µm/hour can be achieved for the deposition of PZT films.

Figure 1. Solmates's Pulsed Laser thin-film Deposition (PLD) system in MESA+ NanoLab's cleanroom (University of Twente, The Netherlands).

In this study, the PZT films were deposited on 4-inch (111)Pt/Ti/SiO$_2$/Si (Pt/Si) and 4-inch (111)Pt/Ti/SiO$_2$/SOI (Pt/SOI, Silicon-on-Insulator) wafers using large-area PLD system with a KrF excimer laser. A 125-nm-thick Pt bottom electrodes and 15-nm-thick Ti adhesive layers were

deposited at room temperature by DC magnetron sputtering on a 500-nm-thick SiO_2 layer formed through wet oxidation at 1100 ^oC. To prevent the formation of pyrochlore phases at the interface between the PZT film and the Pt bottom electrode, a thin nucleation layer of $LaNiO_3$ (LNO, 15 nm, using PLD) was inserted between these layers. The optimized growth conditions for the PZT and LNO layers were: substrate temperature of 600 ^oC; target-substrate distance 60 mm; laser spot size 6.2 mm^2; laser power density 3.0 J/cm^2 and a pressure of 0.1 mbar O_2 during deposition. After deposition, the films were cooled down to room temperature in a 0.1 mbar O_2 atmosphere. Top electrodes (Pt, 125 nm) were deposited on PZT films at room temperature by DC magnetron sputtering.

Crystallographic properties of the PZT films were analyzed with x-ray θ-2θ scans (XRD) using a PANalytical X'Pert X-ray Diffraction system. Film surface and microstructure was investigated using atomic force microscopy (AFM, Bruker Dimension ICON) and high-resolution scanning electron microscopy (HRSEM, Zeiss 1550). The Filmetrics F20 was used to measure the film thickness across the wafer.

For the electrical measurements, 300×300 μm^2 capacitors were patterned with a standard photolithography process and structured by argon-beam etching of the top Pt electrodes and wet-chemical etching (*HF-HCl* solution) of the PZT films to expose the bottom electrodes.

The polarization hysteresis (*P–E*) loop measurements were performed with the ferroelectric mode of the aixACCT TF-2000 Analyzer using a triangular *AC*-electric field of ±200 kV/cm at 1 kHz scanning frequency. All measurements were performed after bipolar cycling the devices a few times. The hysteresis loops did not change after the first few cycles. The longitudinal piezoelectric coefficient (d_{33f}) of the piezoelectric film capacitors was measured with a Double Beam Laser Interferometer (aixDBLI) apparatus that eliminates the influence of the substrate bending[13, 14], using a lock-in technique with a *DC* driving electrical field in the range ±200 kV/cm, and an *AC* peak-peak field amplitude of 5 kV cm^{-1} and 1 kHz frequency. The resolution of the aixDBLI is better than 1 pm/V. In addition, we measured the tip-displacement of 400×100×10 in μm PZT/Si cantilevers to determine the transverse piezoelectric coefficient (e_{31f}). The micro-cantilevers were produced from the PZT films grown on Pt/SOI wafer, and the tip deflection was measured with scanning laser Doppler vibrometer (LDV), driven by the piezoelectric stack at an *AC*-amplitude of ±30 kV/cm (at a *DC* offset voltage of 30 kV/cm) and 8 kHz frequency. The process for fabricating piezoelectric driven Si cantilevers has been described in a previous paper [15]. All measurements were performed at room temperature.
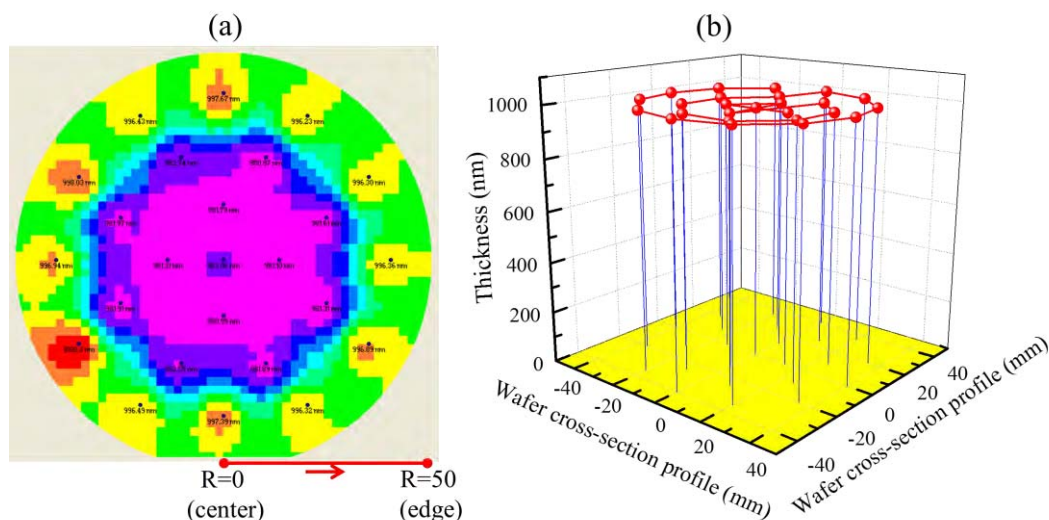

Figure 2. (a) A wafer map of 1-μm-thick PZT film deposited on 4-inch Pt/Si wafer produced by the Filmetrics system and (b) 3D-plot of the thickness uniformity.

3. Results and discussion

The film-thickness uniformity of PZT films on 4-inch Pt/Si wafers was measured using Filmetrics system. Figure 2 shows that the 1-μm-thick PZT films have been formed on 4-inch wafers with thickness variation of less than ±1% (990 nm ± 0.8%) across the wafers. The thickness variation also depends on the film thickness and it slightly increases with thicker films, for instance the 0.5 and 2-μm-thick PZT films have the thickness variation of about 0.4 and 2.2%, respectively.

In Figure 3, the crystallinity of PZT film deposited on 4-inch Pt/Si wafer is given. The XRD patterns at different wafer positions indicate that the relatively good uniformity of crystallinity and the preferred (001) orientation with minor (110) orientation are obtained in the film.
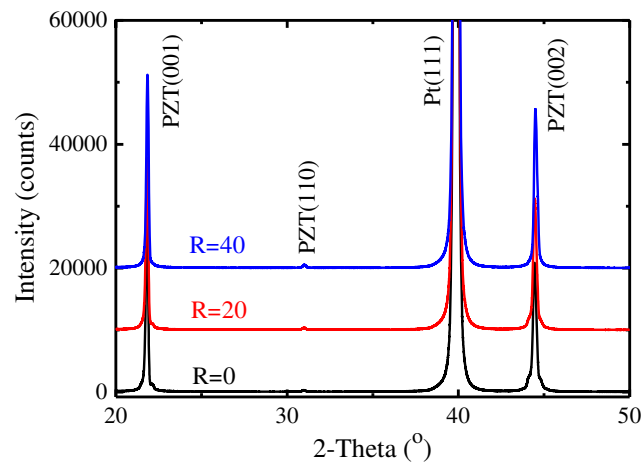
Figure 3. XRD patterns of PZT films at different wafer positions.
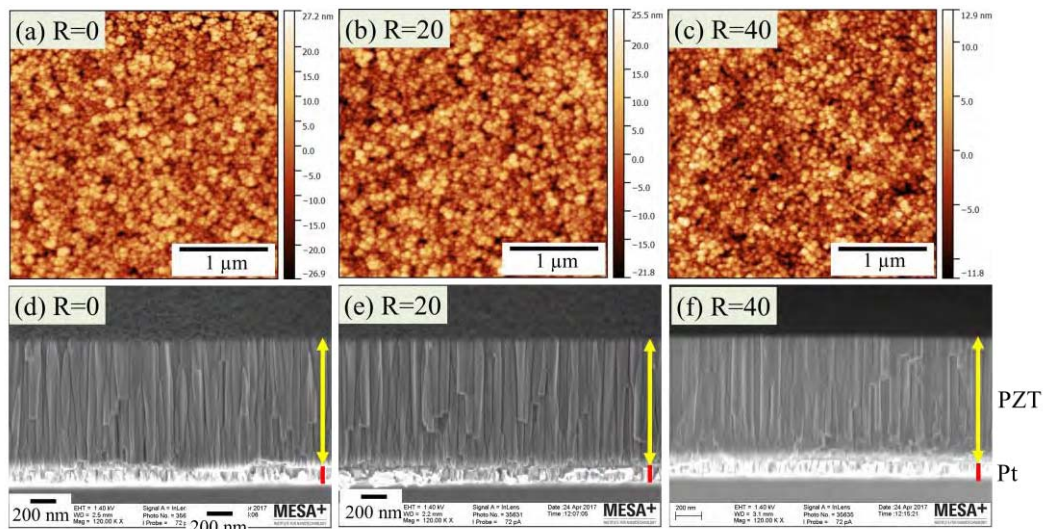

Figure 4. (a-c) Atomic force microscopy (AFM) and (d-f) cross-sectional scanning electron microscopy (SEM) of PZT film deposition on 4-inch Pt/Si wafer, at R=0 mm (wafer center), R=20 mm and R=40 mm (near wafer edge).

The surface morphology and microstructure of PZT films were investigated using atomic force microscopy (AFM) and cross-sectional scanning electron microscopy (SEM) measurements, respectively, as indicated in Figure 4. The columnar grains of PZT films are appeared from the Pt bottom electrodes and the grain size is increases with increasing film thickness. At the topper layer, the grain sizes in the range of 65-75 nm are obtained at the different wafer positions. The root-mean-

square surface roughness (R_{rms}) value, obtained from AFM measurements, is slightly changed from the film at the wafer center (R_{rms} = 7.2 nm) to the film near the edge of wafer (R_{rms} = 4.0 nm). The details are also listed in Table 1. The change in the crystallinity and microstructure of PZT films is probably due to the laser scanning and/or the uniformity of wafer temperature during the deposition.
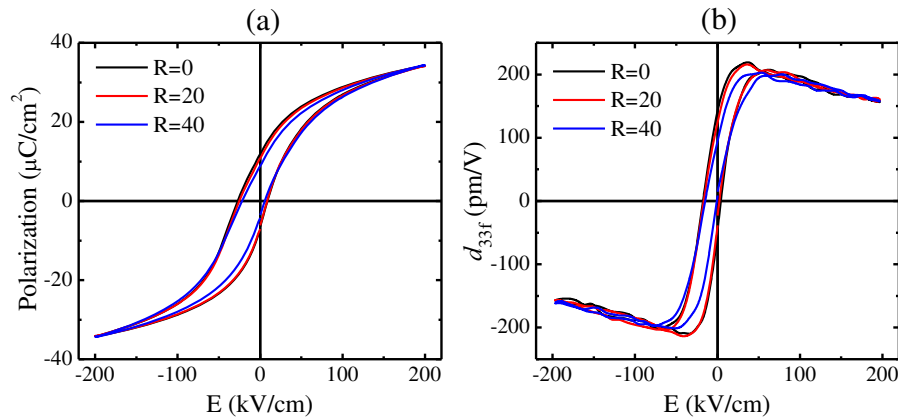

Figure 5. (a) Polarization (*P-E*) and (b) piezoelectric (d_{33f}-*E*) hysteresis loops of PZT films, measured at different wafer positions.
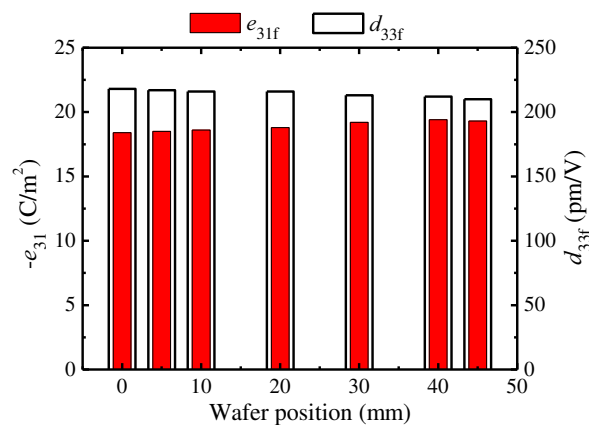
Figure 6. Relationship between piezoelectric e_{31f} and d_{33f} coefficients and position of 4-inch PZT/SOI wafer.

In order to investigate the ferroelectric and piezoelectric properties of PZT films, the polarization (*P-E*) and piezoelectric (d_{33f}-*E*) hysteresis loops were performed at ±200 kV/cm and 1 kHz, as shown in Figure 5. The results indicate that the remanent polarization (P_r), coercive field (E_c) and longitudinal piezoelectric coefficient (d_{33f}), defined from the PZT film capacitors (300×300 μm² in size), are higher at the wafer center. Whereas, the transverse piezoelectric coefficient (e_{31f}) value is slightly enhanced in the devices near the wafer edge (Figure 6). In this case, the more (001) orientation or less (110) orientation in PZT film at the wafer center has a larger P_r value because the spontaneous polarization is along the [001] crystallographic direction. Meanwhile, the enhanced d_{33f} value at wafer center is due to the less closely packed columnar grains, which was previously ascribed to increased declamping of the columns in the film and therefore easier domain wall motion and an increased extrinsic piezoelectric effect [16]. However, the less compacted columnar grain is a possible reason for the lower e_{31f} coefficient from the devices at the wafer center [17].
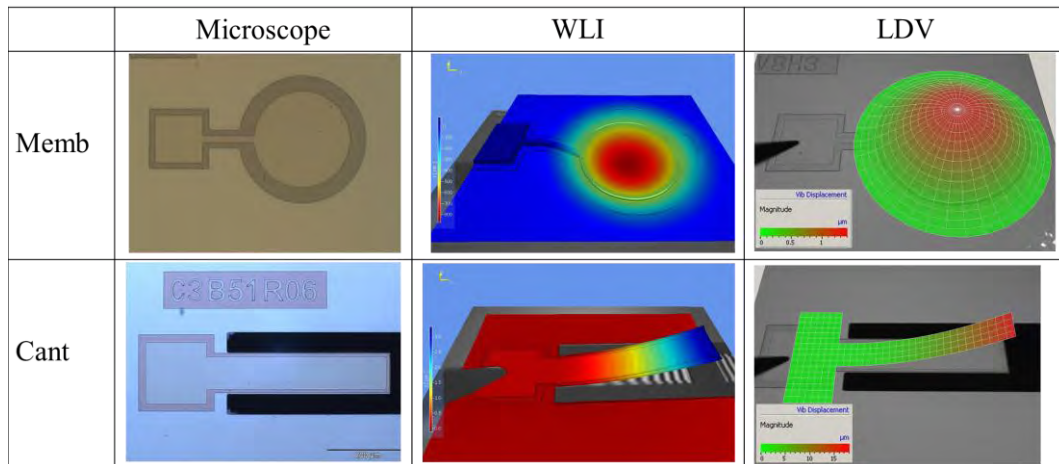
Figure 7. Optical microscope, white-light interferometer (WLI) initial bending and laser Doppler vibrometer (LDV) piezoelectric displacement of a piezoelectric micro-membrane (Memb: 500-µm diameter) and a micro-cantilever (Cant: 400-µm×100-µm, length×width) based on 1-µm-thick PZT films on Pt/10-µm-thick Si device layers.

As mentioned above, there are many potential applications of piezoelectric thin films in the MEMS area. Through large-are PLD technique and wafer-scale microfabrication, the devices based on piezoelectric thin films with different structures can be simultaneously fabricated. Two of these structures, membrane micropumps and resonant micro-cantilever mass-sensors, are shown in Figure 7. The displacement at the center of 500-µm-diameter membrane is about 1.4 µm/V (at the first-mode resonant-frequency of 464 kHz). The quality factor and mass-sensitivity are about 500 and 1 pg/Hz, respectively, for the micro-cantilevers (400-µm×100-µm, length×width) defined at the first-mode resonant peak (with the resonant frequency of 72 kHz). The mass-sensitivity in the cantilevers based PLD films this study is much better than those using the sol-gel (mass-sensitivity: 30.3 pg/Hz) [18] or screen-printing (mass-sensitivity: 62.5 pg/Hz) [19] methods with similar cantilever structure. Moreover, the high yield wafer-scale fabrications of MEMS-based PLD PZT/Si-devices have also obtained. The high uniformity in MEMS devices in the wafer can be seen from the stability in the transverse e_{31f} coefficients where defined from the displacement of cantilevers at different wafer positions (Figure 6).

Table 1. Properties of 1-µm-thick PZT films on 4-inch Pt/Si at different wafer positions.

	Wafer position		
	R=0 mm (center)	R=20 mm	R=40 mm (near edge)
R_{rms} (nm)	7.2	6.5	4.0
P_r (µC/cm^2)	11.8	11.0	9.2
E_c (kV/cm)	18.2	17.1	14.0
d_{33f} (pm/V)	218	216	210
e_{31f} (C/m^2)	-18.4	-18.8	-19.3

4. Conclusion

Piezoelectric PZT thin films have been in-situ deposited on 4-inch Pt/Si or Pt/SOI wafers with large-area pulsed laser deposition (PLD) technique. A high deposition rate of 55 nm/min is obtained. Pure perovskite phase with preferred (001) orientation of the PZT films is achieved. High uniformity in PZT films can be observed from the thickness, microstructure, ferroelectric and piezoelectric properties. The high device yield and reproducibility of high-performance MEMS-based *PLD* PZT/wafer-scales will enhance the applicability of piezoMEMS devices in the industrial applications.

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