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Non-linear piezoelectric properties of the thin $Pb(Zr_xTi_{1-x})O_3$ (PZT) films deposited on the Si-substrate

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Abstract

In this paper some non-linear piezoelectric properties are investigated in lead zirconate-titanate $Pb(Zr_xTi_{1-x})O_3$ (PZT) thin films, sputtered on the $Si/SiO_2/Ti/Pt$ substrates. The thin PZT films were optimised by technology conditions (sputtering (Zr_xTi_{1-x}) composition, PZT film thickness, buffer and seeding layers thickness). The significant piezoelectric response for PZT (60/40) and near MPB PZT (54/46) rhombohedral compositions, (1 1 1) and (0 0 1) orientations and thickness of 1.02–2.2 μm has been observed. The effective piezoelectric coefficient $d_{33} = 225$ pC/N was found for high electric field of 10 MV/m and PZT (60/40) composition. The non-linear piezoelectric response, depending on electric field, frequency and temperature, was studied experimentally using an original double-beam laser interferometer and an optical cryostat. The temperature dependence of the thickness strain was investigated by laser interferometer in the temperature range −33 to 57 °C.

The $Pb(Zr_xTi_{1-x})O_3$-$Si/SiO_2/Ti/Pt$ samples were prepared in the University of Valenciennes (France), and measured in the Laboratory of laser interferometry at the Technical University of Liberec (Czech Republic).

Keywords: Films; Piezoelectric properties; PZT; Actuators; Laser interferometry

1. Introduction

It is well known that the thin and thick lead zirconate-titanate $Pb(Zr_xTi_{1-x})O_3$ (PZT) films are of major interest in the actuation of active structures in micro-electromechanical systems (MEMS) of miniaturised functionality. Because the small thickness of the PZT films results in a high electric field, the non-linear phenomena must be considered both in the design of micro-electromechanical circuit and in the function of the PZT electromechanical system.[1]

To create a Si-based micro-electromechanical system, some technologies as hybrid - or thin film technologies based on the low-pressure chemical vapour deposition (LPCVD), sputtering, and sol gel techniques are used.[2] One of these technologies, i.e. the RF sputtering,[3] was used so that a ferroelectric $Pb(Zr_xTi_{1-x})O_3$ (PZT) thin film on the Si substrate could be obtained.[4,5] The piezoelectric properties of the PZT thin films, namely of (1 1 1) and (1 0 0) orientations,[6] have been extensively studied by many laboratories.[7–9] It is obvious that thin films that are clamped to a substrate, differ from bulk ceramics in microstructure and properties.[10,11]

In the last paper, we described the experimental investigation of the PZT (45/55), PZT (60/40), (1 1 0) and (1 1 1) orientations of the thin films.[12] The aim of this paper is to give an account of the investigation of the piezoelectric response of the $Pb(Zr_xTi_{1-x})O_3$-$Pt/Ti/SiO_2/Si$ structures such as PZT (60/40), (1 1 1) orientation, depending on the technology of preparation, activated electric field, frequency and temperature. The effective piezoelectric coefficients d_{33} of the PZT thin film were determined by piezoelectric induced displacement measured on the thin film sample using the double beam laser interferometer and optical cryostat.[13] The experimentally obtained data are important for verification of some non-linear models of non-linear behaviour of PZT thin film properties.[14]

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2. Samples preparation

The 1.02–2.2 μm thick PZT thin films were grown by RF magnetron sputtering. The targets were cold pressed powders.[3–5] The Zr/Ti ratio was obtained by changing the composition of the targets. The rhombohedral PZT (60/40) thin films were created and used for measurements. The preferential orientation (texture) (1 1 1) was determined with a X-ray diffractometer. The sequence for the (1 1 1) orientation was: PZT (1 1 1)/Pt (100 nm)/TiO_x (20 nm)//SiO_2 (300 nm)/Si substrate (380 μm). On Pt, (1 1 1) orientation was obtained by means of a Pt (1 1 1) bottom electrode and the TiO_2 seeding layer. A gold layer was deposed on the back of the substrate to optimise the laser beam reflection. The PZT films were annealed at 625 °C for 30 min. The parameters of the sputtering are given in Table 1.

A set of measured PZT thin film samples is shortly described in Table 2.

Table 1
Conditions of the sputtering depositions of PZT thin films

Target diameter	76.2 mm
RF power density	2.36 W/cm^2
Target–substrate distance	60 mm
Working pressure	30 mTorr
Substrate temperature	25–160 °C

Table 2
Measured $Pb(Zr_xTi_{1-x})O_3$ thin film samples

Sample	Zr_x	Thickness	Preferentially orientation
#1	0.60	1.0 μm	(1 1 1)
#2	0.60	1.9 μm	(1 1 1)
#3	0.60	2.2 μm	(1 1 1)
#39	0.60	1.7 mm	(1 1 1)
#N3	0.54	1.1 mm	(0 0 1)

Note: C_0 as static capacity measured between top electrodes and bottom electrode varies from 4.11 to 4.78 nF for sample #1. Corresponding permittivity is about 675. The typical diameter of each of top electrodes is 1 mm.
The samples #1, #2 and #3 were delivered in 2003, sample #39 and #N3 in 2002, both from University of Valenciennes (France).

3. Experimental procedure

The piezoelectric-induced thickness displacements of PZT layers were performed with a very sensitive double-beam laser interferometer in the temperature range of −33 to 57 °C (Fig. 1). Based on the He–Ne laser (SIOS model SL03), the Mach–Zehnder double beam interferometer connected to the optical helium cryostat Oxford Instruments, Inc. was designed in our laboratory.[13,15]

To eliminate the vibrations of cooling system caused by the helium pump, our interferometer was miniaturised[15] and situated in the optical head of the cryogenic system. Because the interferometer is very small and compact, the placement inside the optical head of cryostat suppressed the amplitude of vibration of optical elements in the interferometer. Motion of these elements only shifted the interference pattern that remained stable. This technical solution also helped to solve the problem of temperature stability. The thermal shift of optical elements (due to non-homogenous temperature field) and the change of elements length were compensated by the feedback. The interference pattern is transmitted to the photodiode and the signal response from the preamplifier is measured with a digital lock-in (Stanford Research SR-830). The samples in the cryostat were cooled to the chosen temperature and stabilised at this temperature for 10 min. For measurements, the cryostat system was switched off to eliminate sample vibrations of mechanical causes. Under this condition, the piezoelectric response, such as strain, displacement and coefficients d_{33} versus temperature, frequency, and electric field, were measured.

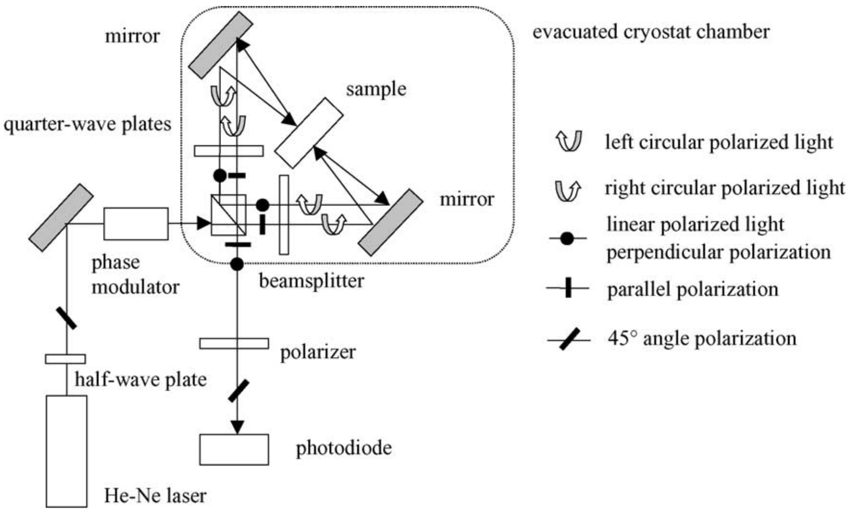

Fig. 1. Schematic drawing of double beam interferometer for measurement inside of helium cryostat chamber (Ref. [13]).

4. Results and discussion

With the aid of the basic formula for thickness displacement Δ ($\Delta = d_{33}\,\mathrm{V_m}$), the effective piezoelectric coefficients d_{33} of all samples were determined after poling them by electric field 20 MV/m within 20 min. In Fig. 2, for rhombohedral PZT (60/40), (1 1 1) sample #39, one can observe a frequency dependence of the piezoelectric response for variable time after poling. Small increase or small changes of the d_{33} (about 120 pC/N) are characteristic for the time delays of 60–1100 min after poling. The frequency of the driving electric field $E_\mathrm{d} = E_\mathrm{m} \sin(\omega t)$ changes from 200 Hz to 2 kHz. The temperature dependence (Fig. 3) of the strain, induced by electric field ($E_0 = 8.31$ MV/m) changes between 160×10^{-6} and 240×10^{-6} (sample #39, 2 min after poling).[12] This value is smaller than the value of the bulk PZT ceramics. The effective coefficient d_{33} decreases with decreasing temperature, but this decrease is not so strong as in case of the bulk samples.[11]

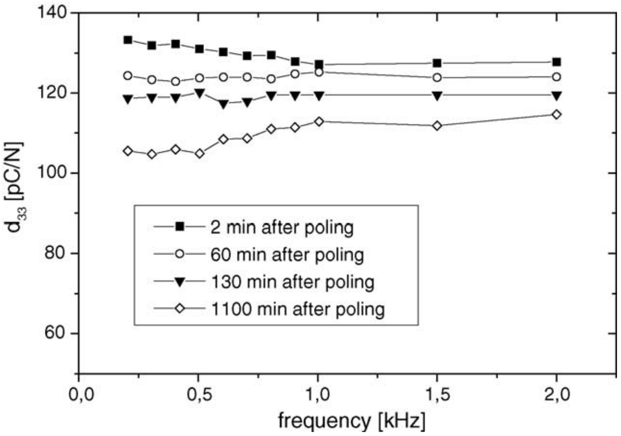

Fig. 2. Frequency dependency of d_{33} coefficient for PZT thin layer, measured at delay times 2, 60, 130 and 1100 min after poling, without bias, sample #39 (Ref. [12]).

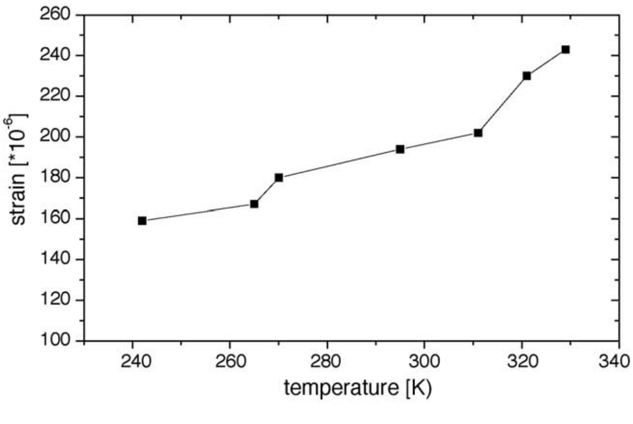

Fig. 3. Temperature dependence of strain for PZT thin layer, frequency driving field 1000 Hz, 10 $\mathrm{V_{ef}}$, sample #39 (Ref. [12]).

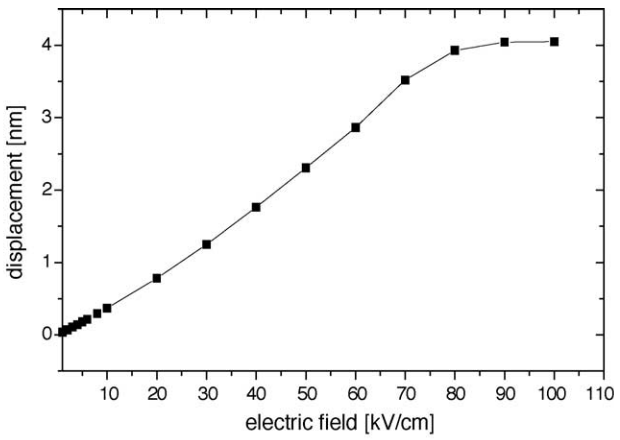

Fig. 4. Piezoelectric induced displacement vs. DC electric field, poled sample #N3.

Fig. 4 shows the non-linear dependence of the thickness displacement (in nm) on varying DC electric field (PZT (54/46), (0 0 1), sample #N3). It is clear that the determination of the threshold field[1] is probably less precise and must be studied in detail. The displacement versus field (and $d_{33}(E)$ dependence) increases quasi-linearly. At higher field (more of 8.0 MV/m) the displacement (and $d_{33}(E)$) becomes constant due to the saturation. The value of electric field, where saturation was observed, is about 8–10 MV/m. These values correspond to the displacements of 4 nm. It is a similar value to the values that have been published before (on 300 nm thin PZT layers[11]). The displacement of 4 nm for sample #N3 correspond to the effective $d_{33} \cong 363$ pC/N. The PZT (60/40) sample # 1 was investigated in high electric fields (0–15 MV/m) and the strain-field dependencies are presented in Figs. 5 and 6. Fig. 5 shows the effective d_{33} coefficient as a function of the applied voltage, sample without poling. The

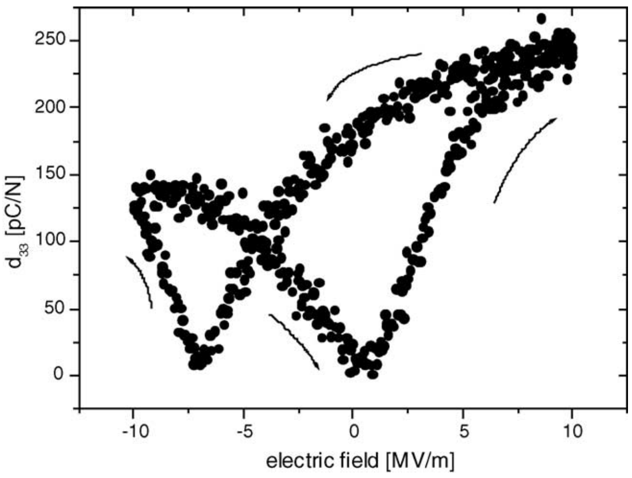

Fig. 5. The effective d_{33} coefficient as a function of the applied voltage, before poling, bias ±10 V of the triangle shape, 1 mHz bias frequency, driving field 1 kHz, amplitude 0.1 MV/m, sample #1.

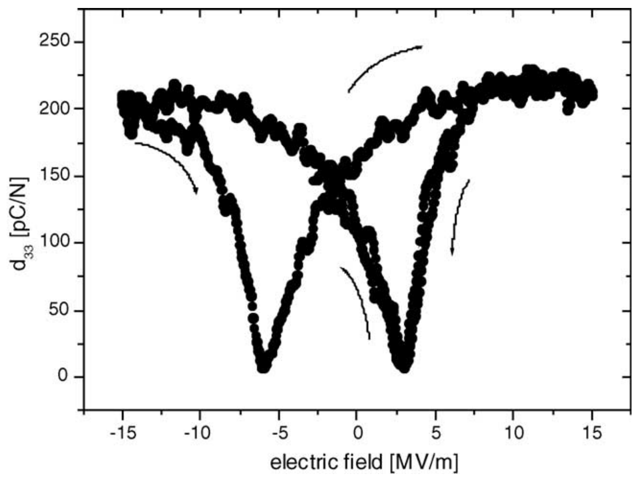

Fig. 6. The effective d_{33} coefficient as a function of the applied voltage, after poling, bias ±15 V of triangle shape, 1 mHz bias frequency, driving field 1 kHz, amplitude 0.1 MV/m, sample #1.

bias voltage is ±10 V of the triangle shape, 1 mHz bias frequency, sample #1.

The influence of the high electric field has been demonstrated. If the applied electric field is higher than the coercive field in the PZT film (averaged coercive field ≈6.8 MV/m for sample #1), ferroelectric domains start to switch. Therefore, the butterfly-type strain hysteresis loop has been observed. This phenomena was reported also by Kholkin et al.[7] Our experiment shows an influence of the poling by a triangle shaped voltage of ±15 V ($E_0 = 8.82$ MV/m). In this case, the typical butterfly-type strain hysteresis loop is quasi-symmetric due to the high electric field, which is significant for poling (Fig. 6). The observed effective piezoelectric coefficient d_{33} is about 225 pC/N. This value is in contradiction with,[8] where the rhombohedral PZT (60/40), (1 1 1) oriented thin film shows smaller values. According to the data published in Ref. 6 for the low applied field, one can observe that interferometer response follows the driving field. This is a case of the pure piezoelectric effect. In low electric field, the strain–field dependencies are quasi-linear. The relative large value of the d_{33} can be produced by large electric field and by local (asymmetric) deformation of the PZT film in the top electrode area under high intensity of electric field (more than 8 MV/m).

This begs an interesting question for a discussion about the possibility of determination both intrinsic and extrinsic contributions to the piezoelectric response of the thin PZT layer.[2]

It is known that created 180° domain wall motion does not involve mechanical strain. Therefore, moving 180° domains contribute only to the dielectric constant ε but not to the d_{33}. We suggest that for ideally (1 1 1) oriented films also the 90° domain contribution does not produce any mechanical strain, which is in agreement with.[1] The non-linear behaviour of d_{33} may give useful information on the relative role of 180° and non-180° domain wall processes in PZT thin films.

5. Conclusions

The technology conditions of rhombohedral PZT (60/40) and PZT (54/46) thin films sputtered on the Si/SiO$_2$/Ti/Pt substrate were optimised so that the (1 1 1) or (0 0 1) texture could be obtained. The non-linear piezoelectric response depending on electric field, frequency, and temperature was studied experimentally using a double beam laser interferometer and an optical cryostat.

Large piezoelectric response as $d_{33} = 225$ pC/N for rhombohedral composition PZT (60/40) was found for abovementioned samples of the thickness 1.7 μm. The temperature dependencies of piezoelectric coefficient d_{33} were measured in the temperature range of −33 to 57°C.

Acknowledgments

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