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Properties of piezoelectric PZT thin films for microactuator applications

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Original version: Materials Research Society Symposium Proceedings 360, 429-434, 1995.
http://hdl.handle.net/10.1557/PROC-360-429

Abstract

The piezoelectric properties of lead zirconate titanate (PZT) thin films deposited on thick silicon substrates and thin silicon membranes were investigated using optical interferometry. The effect of the geometrical constraints and clamping effects on the piezoelectric response is discussed. The study of the dielectric permittivity and the loss as a function of the amplitude of the alternating electric field reveals that extrinsic contributions to the dielectric permittivity become active at large fields. The DC electric field has the effect of freezing out the extrinsic contributions. The influence of the dielectric loss on the piezoelectric properties is discussed.

1. Introduction

The possibility of miniaturization and integration with silicon technology makes piezoelectric thin films interesting for various microactuator devices [1, 2, 3]. In recent years, there was a significant advance in the design and the processing of such devices, which has not been followed, however, by a similar progress in the understanding of the mechanisms which control and affect their piezoelectric response. Due to their essentially two dimensional geometry, thin films exhibit geometrical constraints which must be taken into account during characterization of their properties and interpretation of the data. Because of the small thickness of the films, the effects of the electrodes, the substrate and nonferroelectric layers on the properties cannot be neglected. Due to differences in the processing, the defect structure of the ferroelectric films is expected to be different than in ceramics of the same composition. Thus, the extrinsic contributions to the dielectric, elastic and piezoelectric properties, which are found to dominate the properties of ceramics, may be different in thin films.

In this work, we report and discuss properties of PZT films deposited on thick silicon substrates, and silicon membranes driven by thin PZT films. The films are used for micromotors [4] and various bimorph structures [5]. The piezoelectric properties of the films were investigated by optical interferometry. The effects of clamping of the films on the piezoelectric properties are discussed in detail. In the second part, we present and discuss dielectric permittivity and loss data, measured as a function of alternating (AC) electric field and electric field bias (DC), which demonstrate the presence of extrinsic contributions to the dielectric properties.

2. Preparation of thin film structures

$Pb(Zr_{0.53}Ti_{0.47})O_3$ films on thick silicon substrates (abbreviated herein by FTSS) were prepared using a sol-gel method. The details of preparation are given in [6]. The thickness of the films was 0.3 to l um with top electrodes 0.8 to 2.5 mm in diameter. The wafer was cut into smaller rectangular shaped pieces, with sides typically several millimeters long. Some of the investigated films were doped with Nb. The silicon membranes were micromachined (etched) from silicon wafers, after deposition of electrodes and sol-gel PZT films. The diameter of the membranes was 4 mm, the thickness of the silicon was 10 to 30 um, and the PZT film was 0.6 µm thick. The membranes were used to fabricate micromotors and full description of their preparation, and the configuration and performance of the micromotor can be found in Ref. [4], where results with sputtered PZT films are also presented.

3. Characterization of the piezoelectric properties of films using optical interferometry

Determination of the piezoelectric coefficients of piezoelectric thin films is not a simple task because of the small thickness of the films and their strong clamping to the substrate. The expected displacements due to true piezoelectric effect, even for the electric fields comparable with coercive field, do not exceed l Å. Therefore, optical interferometry is often utilized to determine the longitudinal piezoelectric coefficient, d_{33}, by means of the inverse piezoelectric effect. Up to now, only few results concerning piezoelectric measurements of ferroelectric films have been reported [7-10].

In this investigation, a simple Michelson interferometer [11] was used to measure displacement of FTSS and membranes under external AC electric field. Details of the experimental setup and of the measurement method will be given elsewhere.

The main disadvantage of this experimental scheme is that the displacements of only one face of the sample can be monitored and therefore movement of the whole sample as a rigid body can not be excluded [11]. This problem may be overcome for the bulk samples by clamping the back side of the sample to a rigid holder, and by using samples in a cubic form to prevent the bending effects [12]. In the case of FTSS, the substrate and the film effectively form a bimorph structure resulting in large bending effects even if the frequency is much smaller than the resonance frequency of the flexural mode. Such a behaviour was observed earlier for transverse electroded PLZT films [13]. Under such circumstances, the measured response will not reflect the longitudinal piezoelectric coefficient d_{33}, but rather the transverse coefficient d_{31} which is effectively increased by the multiplication factor of the bimorph. This is illustrated in Fig. 1a and 1b where the displacement of the PZT film on free (unclamped) substrate is plotted as a function of the top electrode diameter and of the distance from the electrode center. The nearly quadratic dependence (Fig. la) and large values of the displacements (Fig. lb) even outside the area of the top electrode, clearly indicate that in this case we are dealing with the movement of the whole substrate. An improvement of the measuring method was achieved by fixing the substrate with rigid silver epoxy exactly under the top electrode. In this case the substrate is allowed to vibrate freely near the edges but is fixed near the point of the probing laser beam. In this way the bending effects were almost eliminated (Fig. 1b).

It should be noted that the direction of the polarization within the films was such to give displacement which was in antiphase with the driving AC voltage in the longitudinal (d_{33}) mode, and in phase for the bending mode which is dominated by the transverse d_{31} coefficient.

Elimination of the bending effects allowed a study of the low-signal piezoelectric response under DC electric field. The hysteresis loop of the piezoelectric coefficient for a Nb doped film is shown in Fig. 2, as an example. The observed butterfly-type loop is common for ferroelectrics with 90° domain wall movement. The loops for positive and negative fields are asymmetric, which is compatible with the measurements of the polarization hysteresis loops. The value of the piezoelectric coefficient at zero field

($d_{33}(0) \sim 70\times10^{-12}$ m/V) should be compared with the values for poled bulk ceramics of the same composition ($d_{33}(0) \sim 70\times10^{-12}$ m/V). The large difference may arise from insufficient poling conditions (films are poled during the short—time hysteresis loop measurements, shown in Fig. 2) and from different micro-structure, domain wall structure and the nature and concentration of defects in ceramics and the

films. Additional data which may offer support to these arguments will be given in the next section. Another reason for the difference certainly arises from the clamping of the film to the substrate. Because of the clamping, the strains along the film surface are not allowed to fully develop and the measured piezoelectric coefficient is reduced, compared to the free material, according to relationship [10]:

$$d_{33}(clamped) = d_{33}(free) - 2d_{31} \cdot s_{13}/(s_{11}+s_{12}),$$

where s_{ij} are the elastic compliances at constant field. Using typical values of the elastic compliances for bulk PZT, and ratio $d_{33}/d_{31} = -2.3$ one obtains that d_{33} measured on clamped thin films may be as small as 50% of the value for the bulk [10, 14].

The amplitude and phase of the interferometric signal are found to be frequency dependent (Fig. 3). The amplitude is slightly decreasing with frequency and is accompanied by a relatively large change of the phase (about 15° from 1 to 100 kHz). A similar behaviour was reported earlier [9] and was related to the influence of residual stresses and increase of the holding force near the film-substrate interface. An alternative explanation is that with increasing frequency the influence of bending vibrations becomes larger, resulting in a decrease of real (due to contributions of the opposite sign from bending, Caused by (d_{31} mode) and increase of imaginary component of the apparent piezoelectric response (due to increase in phase angle as the resonance is approached). Other explanations for the frequency dependence of the apparent piezoelectric response, related to the properties of the material itself, must also be considered.

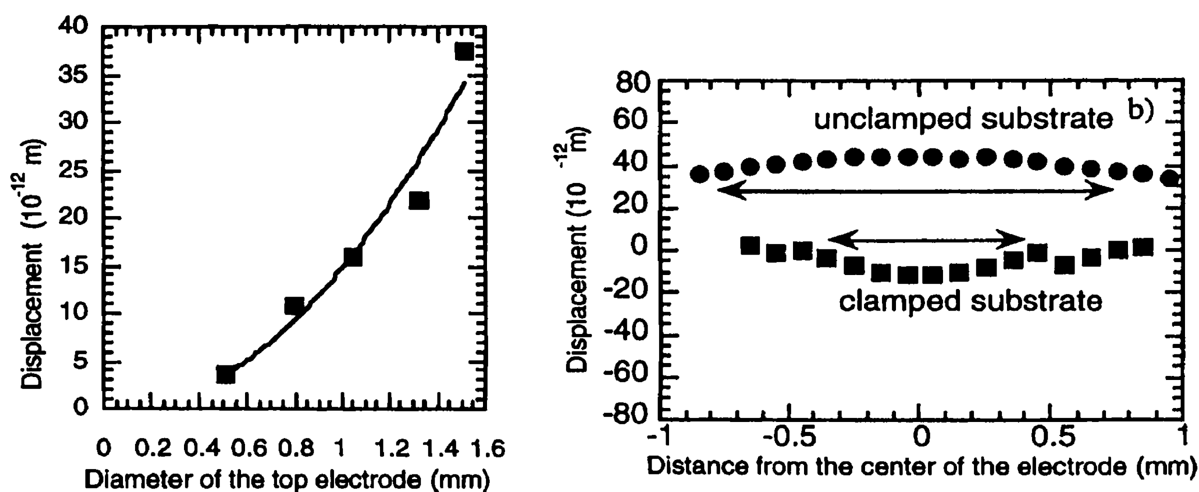

Fig. 1. a) Displacement of the PZT film on unclamped substrate as a function of top electrode diameter. b) Displacement of the PZT film on unclamped and clamped substrate as a function of distance from the center of the top electrode. The arrows indicate electrode width. The frequency of the AC field was 1 kHz, with amplitude 0.1 V for the free and 0.2 V for the clamped substrate.

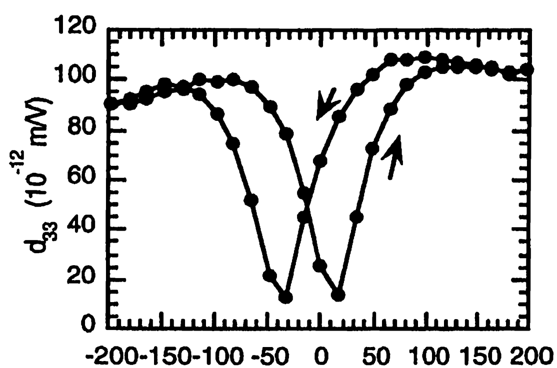

Fig. 2. Hysteresis loop of the piezoelectric coefficient for a 1% Nb-doped PZT film. Frequency of the AC flew was 1 kHz.

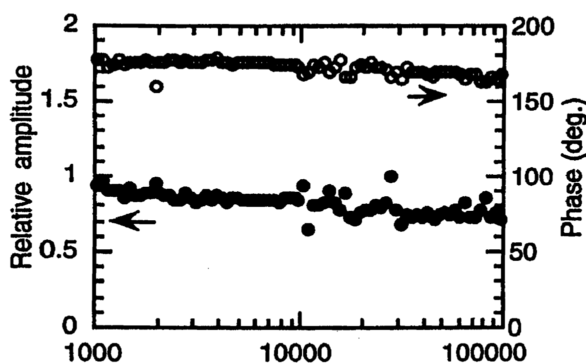

Fig.3. Frequency dependence of the amplitude _ and the phase of apparent piezoelectric response of PZT film on clamped substrate.

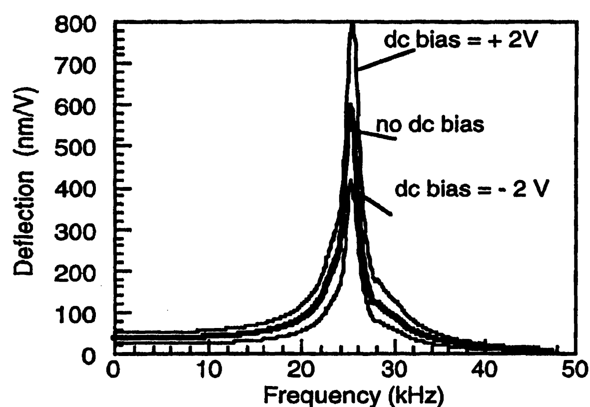

Fig. 4. Deflection of a silicon membrane covered with 0.6 μm thick PZT film.

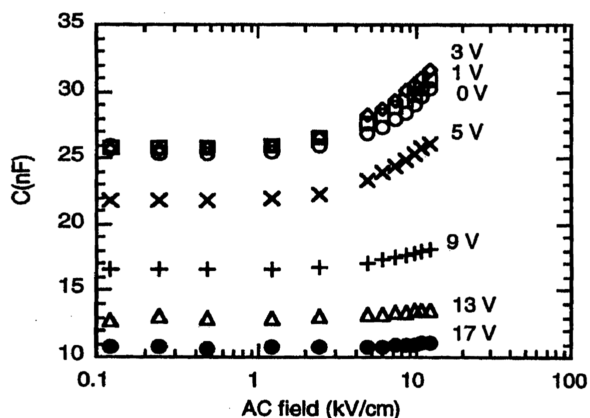

Fig. 5. Fig. 5. Capacitance vs. amplitude of the AC electric field for a sol-gel PZT film deposited on thick silicon substrate. Frequency of AC field was 1 kHz. Diameter of the top electrode was 1.5 mm.

Fig. 4 shows deflection of a silicon membrane covered with sol-gel PZT film, as a function of frequency, near the fundamental mode. Measurements were made by the interferometer, probing the center of the membrane. The AC electric field was 50 mV. The effect of the DC electric field bias is also shown. Deflection is significantly larger for positive bias field. This is in accordance with asymmetry of the hysteresis in piezoelectric coefficient, which is similar to that shown in Fig. 2 for FTSS. Clearly, dependence of the piezoelectric response on the magnitude of the applied electric field should be taken into account when designing devices based on the piezoelectric properties of ferroelectric thin films.

4. Dielectric permittivity and loss of PZT thin films as a function of low signal AC electric field

Fig. 5 shows capacitance (*C*) of a 0.8 μm thick sol-gel PZT film as a function of AC electric field, with DC electric bias field taken as the parameter. Two regions in *C* vs. AC field are obvious: one, at low AC fields, where capacitance is almost constant with field, and the other, above approximately 1 kV/cm, where capacitance increases with the increasing AC field. An analogous behaviour was observed in loss vs. AC field. Similar dependence is reported in PZT ceramics and is interpreted in terms of domain wall motion which is frozen at low excitation fields, but becomes active at large amplitudes [15], where it contributes to the observed dielectric permittivity and loss. Important difference between ceramics and thin films is that the threshold field at which permittivity starts changing, is an order of magnitude higher in PZT films (~l kV/cm) than in ceramics (~0.1 kV/cm) [15]. Possible reasons for the higher threshold field in films are: (i) domains are less mobile in films due to the essentially two dimensional geometry of films (ii) domain walls are pinned by defects which are either not present in ceramics or possess higher activation energy in films than in ceramics, and (iii) the increase in apparent permittivity is not due to moving domain walls, but by some other type of defects or processes. The difference between the threshold fields in the ceramics and in the films reflects similar difference in their coercive fields, which are roughly one order of magnitude higher than the threshold fields.

If domain wall vibrations are responsible for the observed behavior of the permittivity, it may be possible to use the same arguments to explain the comparatively low piezoelectric coefficients in thin films, discussed in the previous section. It has been shown that in piezoelectric ceramics, domain wall motion may be responsible for as much as 70% of the total observed piezoelectric effect [16]. If, as present data might suggest, mobility of domain walls in films is significantly smaller than in ceramics, one may expect to see lower piezoelectric coefficients in the films.

Fig. 5 also shows that the slope of capacitance vs. AC field decreases with DC bias field, in the region above approximately 5 kV/cm, where domain walls (or other defects) are active. This effect of freezing of the domain walls by the DC bias, makes it more difficult for the weak AC field to move domain walls

against the strong DC field. Fig. 6 plots slope of capacitance vs. AC field (from Fig. 5, slope above ~5 kV/cm AC field) vs. DC bias field. At low DC bias, the slope is constant, because the DC field is smaller than the amplitude of the AC field, and DC field cannot freeze the domain walls. The slope starts changing when DC and AC fields become comparable, and domain walls are becoming locked by the DC field.

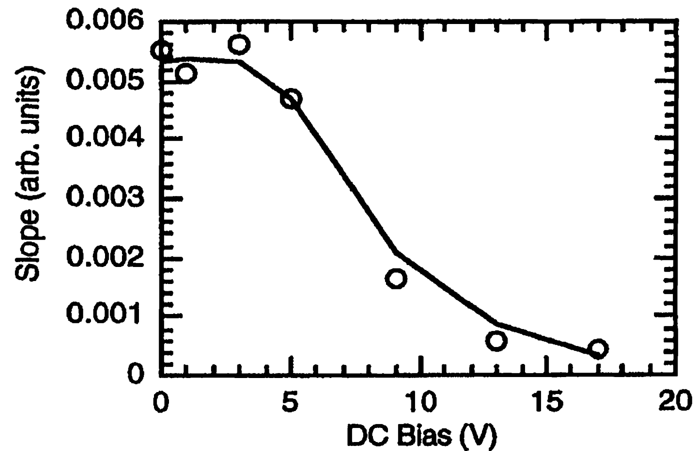

Fig. 6. Slope of capacitance vs. AC field (above 5 kV/cm AC field, Fig. 5) as a function of electric field bias.

5. Acknowledgments

This work was supported in part by the Swiss Federal Office for Science and Education and performed within the European research program on ferroelectric thin films (COST 514).

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