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Electrode size dependence of piezoelectric response of lead zirconate titanate thin films measured by double beam laser interferometry

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The electrode size dependence of the effective large signal piezoelectric response coefficient ($d_{33,f}$) of lead zirconate titanate (PZT) thin films is investigated by using double beam laser interferometer measurements and finite element modeling. The experimentally observed electrode size dependence is shown to arise from a contribution from the substrate. The intrinsic PZT contribution to $d_{33,f}$ is independent of electrode size and is equal to the theoretical value derived assuming a rigid substrate. The substrate contribution is strongly dependent on the relative size of the electrode with respect to the substrate thickness. For electrode sizes larger than the substrate thickness, the substrate contribution is positive and for electrode sizes smaller than the substrate thickness, the substrate contribution is negative. In the case of silicon substrates, if the electrode size is equal to the substrate thickness, the substrate contribution vanishes, and the measured value of $d_{33,f}$ is equal to the theoretical value under the rigid substrate assumption. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4821948]

Piezoelectric and ferroelectric thin films are used in a number of electro-mechanical systems including sensors and actuators. Accurate knowledge of the piezoelectric material properties is crucial for a successful design of such systems. Double Beam Laser Interferometry (DBLI) is a proven, powerful technique for the measurement of the out-of-plane piezoelectric coefficient.[1–3] Nevertheless, since the film is clamped by the substrate, the measured effective piezoelectric coefficient ($d_{33,f}$) is related to both the out-of-plane (d_{33}) and in-plane (d_{31}) piezoelectric coefficients in addition to the elastic properties of the film and the substrate.

A simple theoretical model for $d_{33,f}$ was developed by Lefki and Dormans by assuming completely rigid clamping of the film by the substrate.[4] Considering the elastic nature of the substrate, the clamping by the substrate is never completely rigid. The influence of the elastic properties of the substrate on the value of $d_{33,f}$ has been studied by finite element model simulations.[5] Moreover, the measured $d_{33,f}$ has been shown to have a strong dependence on the top electrode size.[6] Even though it has been recognized that the elastic nature of the substrate plays a role in the electrode size effect,[7] the exact mechanism of this influence has not been well understood. It is generally believed that the mechanics of the layered structure of the film and the substrate is responsible for the observed electrode size effect on $d_{33,f}$. It is also commonly assumed that the saturated value of $d_{33,f}$ vs. electrode size is equal to the true value of $d_{33,f}$.

In this paper, we present a detailed investigation of the mechanism of interaction between the substrate and the piezoelectric thin film that is responsible for the electrode size effect using measurements and finite element modeling. Our study clearly shows that the electrode size dependence of $d_{33,f}$ is primarily due to the substrate contribution. The stresses induced in the substrate due to the mechanical interaction between the piezoelectric film and the substrate cause a small thickness change in the substrate that adds to the expansion of the piezoelectric film when an electric field is applied across the film. Furthermore, we show that this effect is a strong function of the relative size of the electrode with respect to the substrate thickness. In fact, the theoretical value of $d_{33,f}$ defined under the rigid substrate assumption is obtained when the electrode size is approximately equal to the substrate thickness for silicon substrates.

The samples used in this study are lead zirconate titanate (PZT) thin films deposited using a sol-gel process on 8″ Si substrates and are approximately 1.9 μm thick. The bottom electrode is 100 nm thick platinum and the top electrode is 100 nm thick ruthenium. Top electrodes were defined by photolithography and consisted of both square and circular contacts with sizes ranging from 0.1 mm to 2 mm. An aixACCT DBLI system (model: aixDBLI industrial line) was used to measure small and large signal $d_{33,f}$ at different field strengths, although only large signal measurement results are presented here. Hence, in this work, we label our measurement results as $d_{33,f,ls}$ to distinguish them from the simulation results which are labeled as $d_{33,f}$. The samples exhibit extremely uniform piezoelectric properties across the 8″ wafer with an average measured large signal ($d_{33,f,ls}$) value of 185 pm/V and a standard deviation of 4.5 pm/V at 150 kV/cm for 2 mm × 2 mm square electrodes (based on measurements at 43 different locations across the wafer).

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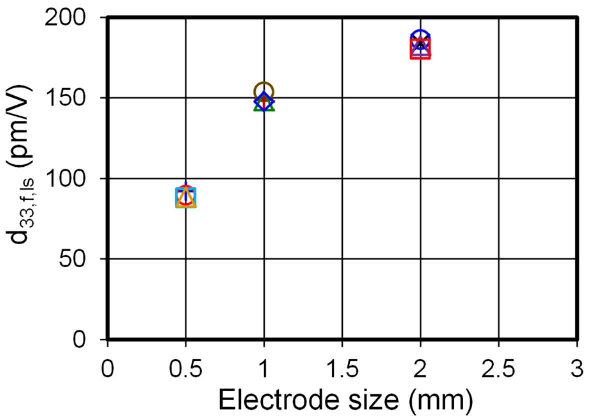

FIG. 1. Measured electrode size dependence of $d_{33,f,ls}$. Substrate thickness = 725 μm. The different symbols correspond to measurements on different wafers and different locations. Overlapping of multiple symbols shows the excellent uniformity of the PZT properties across the wafer and from wafer to wafer.

The electrode size dependence of $d_{33,f,ls}$ is shown in Fig. 1 and it is similar to the results reported in the literature.[6,7] Note that these data are from two different locations from two different wafers. This illustrates the high level of PZT uniformity not only across the wafer but between wafers. Such uniformity gives us confidence to relate the $d_{33,f,ls}$ variations solely to the electrode size and not to the non-uniformity of PZT.

To understand the electrode size dependence, 3D finite element modeling was performed using COMSOL 4.2. The simulated structure consists of a sample 1.9 μm thick PZT film on a 725 μm thick Si substrate as shown in Fig. 2. Taking advantage of the four-fold symmetry, only a quarter structure was modeled. Some of the material properties used for modeling are summarized in Table I. Unless a specific material property value is varied for investigation as noted in the "variants" column in Table I, nominal values of all the other parameters are used in the simulation.

In the model, the electrode size was varied from 0.1×0.1 mm^2 to 2×2 mm^2 and the substrate/film size was fixed at 8×8 mm^2. Larger size substrates (e.g., 16×16 mm^2) were also modeled but gave nearly identical results for $d_{33,f}$. The substrate thickness was varied in the range of 100 μm to 725 μm. The effects of varying the elastic properties (Young's modulus and Poisson's ratio) of the PZT film and of the substrate, as well as the piezoelectric coupling coefficients d_{33}, d_{31}, and their ratios were also investigated.

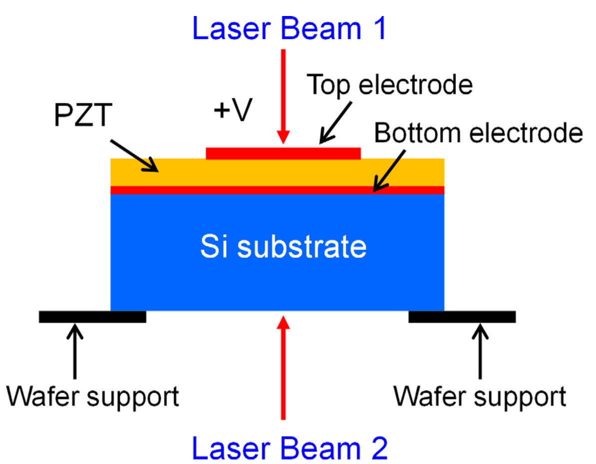

FIG. 2. Schematic diagram of a typical DBLI measurement set-up including the cross-section of a simulated structure.

TABLE I. Material properties used for modeling.

Property	Nominal	Variants
Si Young's modulus	150 GPa	110, 150, 200 GPa
Si Poisson's ratio	0.3	0.25, 0.3, 0.35, 0.4
PZT Young's modulus	60 GPa	60, 70, 80, 90, 100 GPa
PZT Poisson's ratio	0.3	0.25, 0.3, 0.35, 0.4
PZT thickness	1.9 μm	
Si substrate thickness	725 μm	100, 225, 350, 475, 600, 725 μm
d_{33}	300 pm/V	200, 300 pm/V
d_{31}	−150 pm/V	−100, −120, −140, −160 −180 pm/V

In order to make a meaningful comparison between modeling and the measurement results, it is very important to use realistic boundary conditions that closely represent the actual measurement conditions. A schematic representation of the DBLI measurement is also shown in Fig. 2. This measurement technique uses two vertically aligned laser beams, one focused at the middle of the top electrode and the other focused on the bottom polished surface of the Si substrate. The sample sits on a stage with a large hole which allows the passage of the bottom laser beam. Under these experimental conditions, when an electric field is applied across the PZT film, the sample is free to bend about the boundary lines at the bottom of the substrate that are aligned with the edges of the hole in the sample stage. Hence, our choice of the mechanical boundary condition for all the simulations reported here is to fix the bottom of the substrate along these boundary lines. We also investigated other types of mechanical fixed boundary conditions and a detailed account of the results for these boundary conditions will be presented elsewhere.

The Z-direction (vertical) displacement of the points aligned with the center of the electrode on the top surface of PZT, bottom surface of PZT, and the bottom surface of the Si substrate were extracted from the simulated model. The difference in Z-displacement between the top of the PZT film and the bottom of the Si substrate per unit applied voltage gives $d_{33,f}$ as measured by a physical DBLI system. The value of $d_{33,f}$ simulated with these conditions (delta_tot) is plotted in Fig. 3 as a function of electrode size. It is seen that $d_{33,f}$ increases with increasing electrode size and saturates at a constant value for large electrode sizes, consistent with the measurement results shown in Fig. 1. In addition, the model leads to an interesting result that cannot be directly observed in measurements. The difference in Z-displacement between the top and bottom of the PZT thin film (delta_PZT) is independent of the electrode size. Interestingly, the PZT contribution to $d_{33,f}$ is found to be exactly equal to the theoretical value derived by Lefki and Dormans[4] under the rigid substrate assumption

$$d_{33,f} = d_{33} - \frac{2 d_{31} s_{13}}{(s_{11} + s_{12})}, \tag{1}$$

where d_{33} and d_{31} are the true piezoelectric constants, and s_{11}, s_{12}, and s_{13}, are the elastic compliance coefficients of the

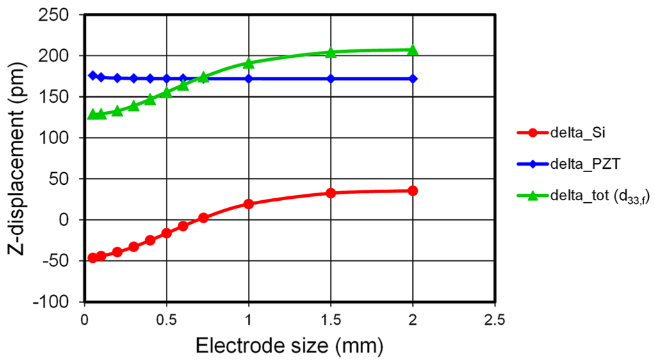

FIG. 3. Thickness change in Si, PZT, and the total ($d_{33,f}$) for 1 V as a function electrode size.

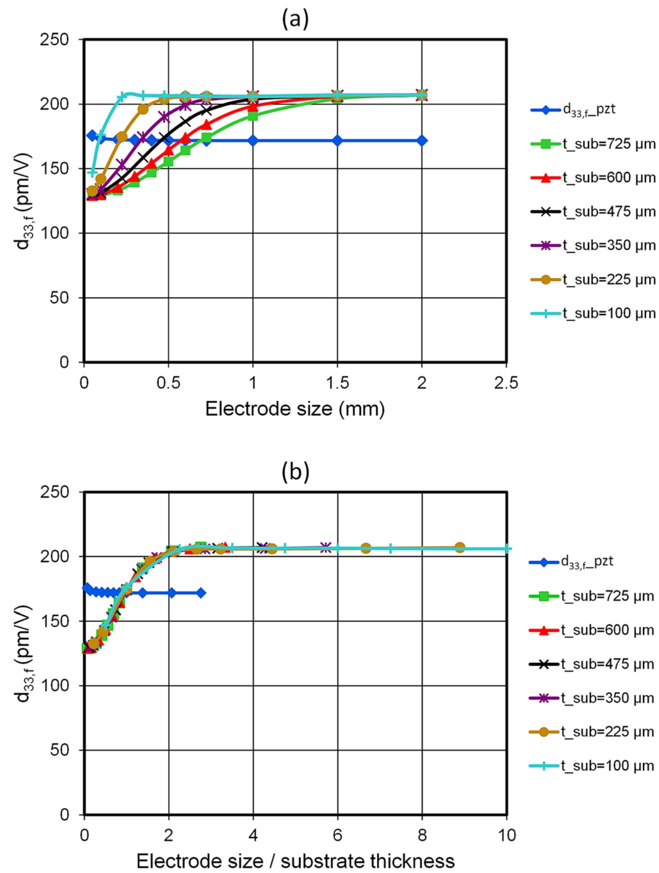

FIG. 4. Simulated $d_{33,f}$ for different substrate thicknesses (a) as a function of electrode size and (b) as a function of electrode size/substrate thickness.

piezoelectric material. This also implies that the electrode size effect on $d_{33,f}$ is actually due to the substrate contribution (delta_Si) arising from the mechanical interaction between the PZT and the substrate when an electric field is applied across the PZT. For large electrode sizes, delta_Si is positive and reaches a constant value while for small electrode sizes it is negative. Additionally, the delta_Si curve crosses zero when the electrode size is approximately equal to the thickness of the substrate. Equivalently, the theoretical value of $d_{33,f}$ under the rigid substrate assumption is obtained when the electrode size is equal to the thickness of the substrate. For larger size electrodes, the substrate contribution is positive and the measured value of $d_{33,f}$ is larger than the theoretical value while for electrode sizes smaller than the substrate thickness, the substrate contribution is negative and the measured value of $d_{33,f}$ is smaller than the theoretical value. It is important to note that the saturation value of $d_{33,f}$ observed when the electrode size is $\geq$3x substrate thickness is actually larger than the theoretical value derived under the rigid substrate assumption.

To verify if the intersection between delta_tot and delta_PZT always occurs when the electrode size is approximately equal to the Si substrate thickness, we performed several simulations with different substrate thicknesses and the results are shown in Fig. 4(a). Since the PZT properties are the same for all the substrate thickness, there is only one constant d_{33,f_pzt} curve (equal to delta_PZT). For different substrate thickness, the total $d_{33,f}$ curves (delta_tot) shift and in every case the crossing is found to occur when the electrode size is equal to the substrate thickness. The same data are replotted in Fig. 4(b) with the ratio of electrode thickness to the substrate thickness as abscissa, and, in fact, all the curves for different substrate thickness collapse into a single curve.

To validate our modeling prediction, we also performed large signal $d_{33,f,ls}$ measurements as a function of electrode size (for both square and circular electrodes) for different substrate thicknesses and the results are shown in Fig. 5. Indeed, the data points for different substrate thickness collapse into a single curve, as predicted by our modeling and the true value of $d_{33,f,ls}$ is given by the value at electrode size/substrate thickness equal to unity (which in our case is approximately 110 pm/V).

The physical mechanism responsible for the substrate contribution to $d_{33,f}$ can be understood by considering the lateral (in-plane) and the vertical (out-of-plane) substrate stresses caused by the mechanical interaction between the electrically activated PZT and the substrate. For large electrode sizes (>substrate thickness), our modeling shows that there is considerable bending of the structure and the out-of-plane stress in the substrate is nearly zero. However, there is substantial in-plane compressive stress in the substrate due to the lateral squeezing of the substrate by the PZT film resulting in an expansion of the substrate thickness (positive contribution to $d_{33,f}$). On the other hand, for smaller electrode sizes (<substrate thickness), the bending of the substrate is very small and the PZT pushes down the substrate at the PZT/substrate interface. This produces a large out-of-plane compressive stress in the substrate causing the substrate thickness to shrink (negative contribution to $d_{33,f}$). For intermediate electrode sizes, the net substrate effect is determined by the relative contribution from the in-plane compressive stress (expansion) and the out-of-plane compressive stress (squeezing). It turns out that in the case of Si substrates, the two contributions cancel out each other when the electrode size is nearly equal to the substrate thickness, and the measured value of $d_{33,f}$ is equal to the theoretical value derived under the rigid substrate assumption.

It is worth noting that the DBLI measurement technique eliminates any bending contribution to $d_{33,f}$ through the use of dual laser beams.[1] In bending, the vertical displacements can be very large (several times the PZT expansion), but the top and bottom surfaces move together. However, in the case of stress-induced change in substrate thickness, there is

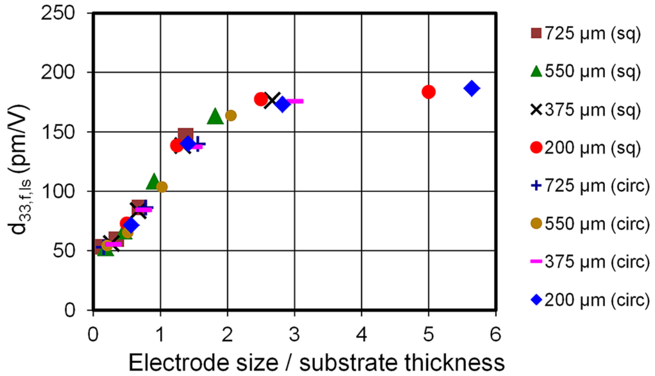

FIG. 5. Measured large signal $d_{33,f,ls}$ for different substrate thicknesses as a function of electrode size/substrate thickness.

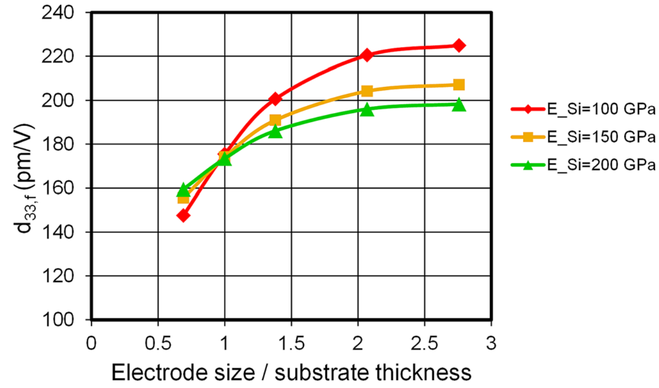

FIG. 6. Simulated $d_{33,f}$ for different substrate Young's modulus as a function of electrode size/substrate thickness.

relative motion between the top and bottom surfaces adding to the value of $d_{33,f}$ measured by DBLI.

We also performed simulations for various values of PZT Young's modulus, PZT Poisson's ratio, and different combinations of d_{33} and d_{31} and it is found that the value of $d_{33,f}$ that would be measured in a DBLI measurement is equal to delta_PZT (theoretical $d_{33,f}$ for rigid substrate) in all the cases when the ratio of the electrode size to substrate thickness is approximately unity in the case of Si substrates. More detailed results of this investigation will be presented elsewhere. Hence, we believe that our process for measuring the true $d_{33,f}$ using an electrode size equal to the substrate thickness is valid not just for PZT but in general for any piezoelectric material. In fact, our measurement and modeling results on AlN indeed validate this claim and the results will be published elsewhere.

We also investigated the influence of the substrate elastic properties on the electrode size dependence of $d_{33,f}$ and the results are shown in Fig. 6. It is seen that the curves get flatter and the saturation value of $d_{33,f}$ gets closer to the theoretical $d_{33,f}$ as the substrate modulus is increased. Furthermore, all the curves cross when the x-axis value (the ratio of electrode size to the substrate thickness) is unity. In the limit of a fully rigid (infinite modulus) substrate, the line will be completely flat (not shown in Fig. 6) going through the same intersection point. Thus, even though the measured value of $d_{33,f}$ depends on the substrate modulus, as reported by Prume *et al.*,[7] the reference ratio at which the measured $d_{33,f}$ is equal to the theoretical $d_{33,f}$ (for rigid substrate) turns out to be unity, independent of substrate Young's modulus. However, the substrate Poisson's ratio is found to affect this reference ratio. Though the physical reason for this dependency is not completely understood, we believe that the substrate Poisson's ratio affects the interplay between the vertical and the lateral stresses in the substrate, and hence alters the reference ratio. This will be investigated further in a future publication. Nevertheless, there is a fixed unique ratio of electrode size to substrate thickness (depending on the substrate Poisson's ratio) at which the measured value of $d_{33,f}$ is equal to the theoretical value of $d_{33,f}$ for a rigid substrate.

In conclusion, we have investigated the electrode size dependence of the piezoelectric response coefficient ($d_{33,f}$) of thin films using both DBLI measurements and COMSOL 4.2 modeling. Our investigations clearly show that the observed electrode size dependence of $d_{33,f}$ is purely due to the substrate contribution. The PZT contribution to $d_{33,f}$ is shown to be independent of electrode size and is equal to the theoretical value derived assuming a rigid substrate. The substrate contribution is a strong function of the relative size of the electrode with respect to substrate thickness and this contribution vanishes when the ratio of electrode size to substrate thickness is unity in the case of Si substrates. The ratio of the electrode size to the substrate thickness at which the substrate contribution vanishes is independent of the PZT properties and substrate Young's modulus but depends on the substrate Poisson's ratio.

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