Effects of Elastic Coupling between $BaTiO_3$ Ferroelectric Film and a Substrate with Finite Thickness on Piezoelectric Coefficients

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The effective piezoelectric coefficients of $BaTiO_3$ ferroelectric films epitaxially grown on different single crystal substrates with finite thickness have been theoretically analyzed. The effective longitudinal converse piezoelectric coefficients d_{33} of film and "film–substrate" heterostructure all monotonously increased with increase of the film thickness fraction k, and the latter is always larger than the former at the range of $0 < k < 1$. Meanwhile, we also found that the effective piezoelectric coefficients d_{33} were affected by the substrates due to different elastic constants. These results show that the elastic deformation and clamping effect of substrate have significant impacts on the piezoelectric behavior of bilayer heterostructure.

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1. Introduction

Piezoelectric materials can be used as sensors, where the application of mechanical strains or stresses produces an electric charge signal, or actuators, where an applied field induces mechanical strain [1–5]. It is well known that the above electromechanical coupling behaviors can be characterized by longitudinal piezoelectric coefficients in evaluating the piezoelectric performance. The accurate measurement of piezoelectric coefficients has been achieved in bulk materials for a long time. However, in piezoelectric films, there is a requirement to measure longitudinal piezoelectric coefficients on 2D film–substrate heterostructure systems. Thus the measurement will be influenced inevitably by the substrate, including the substrate clamping effect, substrate deformation, etc. They should be carefully considered as well as the measurement manner when studying the piezoelectric properties of multilayer heterostructure.

With the trends in miniaturization, it is expected that piezoelectric film heterostructures with their excellent electromechanical coupling should and will play an important role in microelectromechanical system (MEMS), even in nanoelectromechanical systems (NEMS) [6, 7]. Different from corresponding bulk materials, many film heterostructures have complex geometries in microdevices which require using numerical analysis methods to evaluate performance, but it is difficult to be undertaken for such analyses without comprehensive understanding of the full set of properties of the films. These considerations have promoted to understand the piezoelectric performances of the film heterostructures [8–11]. Lefki and Dormans proposed two methods for treating a ferroelectric film [12]. Measurement of the charge/voltage response with a uniform stress normal to the film surface and measurement of the change in thickness of the film with an electric field applied to the electrodes. In the latter case, ferroelectric film is exposed to an out-of-plane electric field (polarization is not totally aligned in-plane), it will deform along the electric field in the mode of converse longitudinal piezoelectric d_{33}^{f}, where the superscript "f" denotes "film". It was assumed that

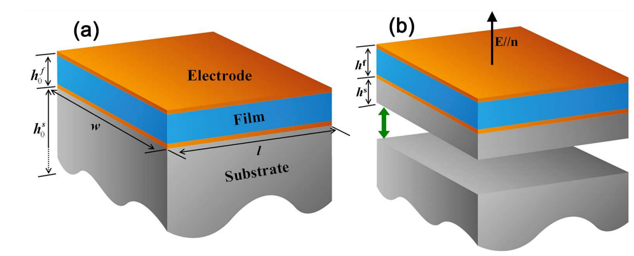

Fig. 1. Schematic of a heterostructure: (a) the substrate is absolutely rigid and much thicker than the film, (b) the substrate with a finite rigidity is microfabricated to be thin in some devices.

the substrate is absolutely rigid and much thicker than the ferroelectric film here (see Fig. 1a), so an approximation can be obtained in heterostructure that the piezoelectric film is perfectly clamped by the underlying substrate. Thus the elastic constraint from the substrate

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creates in-plane stresses in the film, which reduces the out-of-plane piezoelectric deformation of the film via the Poisson effect [12]. With the Lefki–Dormans' formulation [12–14], the effective converse longitudinal piezoelectric coefficient for a $(00l)$-oriented ferroelectric film with a tetragonal/pseudo-tetragonal structure is given by

$$d_{33}^{f} = \frac{\varepsilon_{33}}{E_3} = d_{33} - \frac{2S_{13}^{f} d_{31}}{S_{11}^{f} + S_{12}^{f}}.$$

The same Z-value will be obtained regardless measured by total surface displacement ($d_{33,d}^{f}$) or by only film strain ($d_{33,\varepsilon}^{f}$) here. d_{ij} and S_{ij} $(i, j = 1,\ 2,\ 3)$ are the piezoelectric and elastic compliance tensors of the corresponding bulk samples, and ε_{ij} are the piezoelectric strain tensors. Throughout the paper, the elastic compliances and moduli refer to those measured at a constant electric field.

However, the piezo-strain of a constrained film cannot be directly measured by the standard resonance method due to the substrate clamping. In the above case, the effect of the substrate deformation is eliminated by probing the heterostructure on both the film side and on the back side of the substrate. But if the substrate is not absolutely rigid, even if a film is thinner than the substrate, the total displacement still depends on the elastic properties of the substrate. So, with the direct measurement method, we cannot ignore the deformation of the substrate due to the applied pressure. Meanwhile, the bending effect of the heterostructure also has some contribution to the piezoelectric response and should be considered in evaluating the piezoelectric performance. The suppression methods for bending deformation have been well study in the literatures [14–17]. The effective converse longitudinal piezoelectric coefficient measured through the total piezo-response of a $(00l)$-oriented film-substrate heterostructure can be obtained with the formula [14, 16]:

$$d_{33,d}^{f} = d_{33} - \frac{2s_{13}^{f} d_{31}}{s_{11}^{f} + s_{12}^{f}} + \frac{2s_{13}^{s} d_{31}}{s_{11}^{f} + s_{12}^{f}}.$$

The last term on the right side is an additional positive contribution due to the elastic deformation of the substrate because the compliance S_{13}^{s} has a negative value generally.

The above analysis is only valid for the case shown in Fig. 1a, where the thickness of film is much smaller than the substrate. But in some devices, a substrate with a finite rigidity is microfabricated to be thin (see Fig. 1b), i.e. the thickness of the substrate may be much thinner than the film, or even deliberately etched out to leave films only ($h^s \to 0$ in Fig. 1b). To allow one to compute the effective longitudinal coefficients for arbitrary thickness, the objective of this paper is to provide a more general formulation, where the thickness fraction of the film in the heterostructure, the elastic properties and piezoelectric properties of the film and the substrate were taken into account, for predicting the deformation of a film–substrate heterostructure which relaxes assumptions made in previous models. In addition, the paper will highlight the effect of elastic properties of the substrate on piezo-deformation of the film and heterostructure.

2. Theoretical analysis

As shown in Fig. 1b, the electrical field is applied along the thickness axis n of a film–substrate heterostructure (the film normal is denoted by z). The thicknesses of the ferroelectric film and substrate are denoted by h^f and h^s, respectively. Assuming that the total thickness of the heterostructure ($h = h^f + h^s$) is much smaller than the in-plane dimensions ($= l$ and w), the elastic problem becomes one-dimensional. The bending contribution of the heterostructure is eliminated in the present case, so the internal stress is given by $\sigma_{lm}(z) = G_{lmpq}[\overline{\varepsilon}_{pq} - \varepsilon_{pq}^{0}(z)]$ [18]. On the other hand, the average internal stress of heterostructure should be equal to zero, which can be represented by an integral equation

$$\int_0^h G_{lmpq}[\overline{\varepsilon}_{pq} - \varepsilon_{pq}^{0}(z)]\,\mathrm{d}z = 0, \tag{1}$$

where $\overline{\varepsilon}_{pq}$ and $\varepsilon_{pq}^{0}(z)$ are the average strain and location-dependent self-strain tensors, $G_{lmpq} = C_{lmpq} - C_{lmri}n_r\ [n_u C_{uikv} n_v]^{-1} n_t C_{ktpq}$ are planar elastic modulus tensors, n are the normal vectors and C are the elastic moduli.

Then the average strain $\overline{\varepsilon}_{pq}$ can be solved with Eq. (1) based on the input of self-strain $\varepsilon_{pq}^{0}(z)$, and the self-strain is just the piezoelectric strain in the film and zero in the substrate in this case, the piezoelectric strain of the film can be expressed as $\varepsilon_{pq}^{P}(z) = d_{wpq} l_w E$, where d_{wpq} is the piezoelectric coefficient tensor of a bulk ferroelectric, l_w are the direction cosines of the electric field E, and superscript P denotes the piezoelectric strain

$$\varepsilon_{pq}^{0}(z) = \begin{cases} \varepsilon_{pq}^{P}(z), & h^s < z \le h, \\ 0, & 0 \le z \le h^s. \end{cases} \tag{2}$$

Then the average strain in the film–substrate heterostructure is easily found to be

$$\overline{\varepsilon}_{pq} = k\overline{S}_{lmpq}(k) G_{lmpq}^{f} \varepsilon_{pq}^{P}(z), \tag{3}$$

where $k = h^f/h$ is the thickness fraction of the ferroelectric film, and $\overline{S}(k)$ is the average planar elastic compliance of the film–substrate heterostructure and the corresponding tensor was defined by $\overline{S}_{lmpq}(k) = [kG_{lmpq}^{f} + (1-k)G_{lmpq}^{s}]^{-1}$, G^f and G^s are the planar elastic moduli of the film and substrate, respectively.

The local normalized displacement is given by [18]:

$$\delta_i(z) = [n_u C_{uikv} n_v]^{-1} n_t C_{ktpq} [\varepsilon_{pq}^{0}(z) - \overline{\varepsilon}_{pq}]. \tag{4}$$

Then substituting Eqs. (2) and (3) into Eq. (4), the normalized displacement vectors for the film and substrate can be solved. Meanwhile, the longitudinal coefficients measured by film strain can be obtained by $d_{\mathrm{ln}}^{f} = \delta_i n_i / E$ [15]. When the applied electrical field is along the orientation of the film thickness, the longitudinal piezoelectric coefficient d_{33}^{f} of the film is given by

$$d_{33,\varepsilon}^{f} = [n_u C_{uikv}^{f} n_v]^{-1} C_{ktpq}^{f} d_{wpq} n_i n_t n_w$$

$$\times[1 - k\overline{S}_{lmpq}(k)G^f_{lmpq}]. \quad (5)$$

On the other hand, combining with the contribution of the deformation of the substrate, the apparent piezoelectric coefficients may also be obtained by $d_{\text{ln}} = (\delta^f_i n_i h^f + \delta^s_i n_i h^s)/(h^f E)$, the longitudinal piezoelectric coefficient d^f_{33} of the heterostructure can be defined as the total displacement, which can be written as

$$d^f_{33,d} = d^f_{33,\varepsilon} - (1-k)\overline{S}_{lmpq}(k)[n_u C^s_{uikv} n_v]^{-1}$$
$$\times C^s_{ktpq} d_{wpq} n_i n_t n_w G^f_{lmpq}. \quad (6)$$

If the intrinsic piezoelectric coefficients of the film and the elastic constants of the film and substrate are known, the effective longitudinal coefficients for arbitrary thickness of a film–substrate hetero-epitaxial structure can be computed by the above two Eqs. (5) and (6).

This theoretical analysis is based on the following assumptions:

(i) The bending effect is eliminated by using a simplified "film–substrate–film" tri-layer structure, which is equivalent to a film–substrate bi-layer structure having a fixed bottom surface in terms of piezoelectric responses.

(ii) Electrode layers are perfect conductors and are infinitesimally thin, so they have no mechanical effect on the deformation of the film–substrate heterostructure.

(iii) The film is assumed to be epitaxially grown on the substrate, free of defects and in equilibrium. It should be noted that most films in MEMS applications have a textured polycrystalline microstructure, however, our general solutions are still applicable in these films by using pseudo-tetragonal elastic constants of the corresponding bulk materials.

(iv) The heterostructure temperature is uniform and under no mechanical loading.

(v) Plane dimension of the film is much larger than the total thickness of the heterostructure (l and $w \gg h$) so that edge effects may be neglected.

3. Results and discussion

The piezoelectric properties of $Pb(Zr_xTi_{1-x})O_3$ film have been widely and systematically studied in the literatures [6, 19–21]. However, detailed investigations about lead-free ferroelectric films like $BaTiO_3$ (BTO) film heterostructure are very few. In this paper, longitudinal piezoelectric coefficients $d^f_{33,\varepsilon}$ and $d^f_{33,d}$ of BTO epitaxial film heterostructure were analyzed based on the above theoretical calculations. Si, $SrTiO_3$ (STO), $LaAlO_3$ (LAO), and MgO were employed as single crystal substrates for BTO film growth. The bulk piezoelectric coefficients and the elastic moduli used for BTO materials were taken from Devonshire crystal values [22, 23], and with the elastic compliance coefficients of substrates [24–26] were summarized in Table I.

Take epitaxial $BTO_{(001)}/LAO_{(001)}$ film–substrate heterostructure for example, based on Eqs. (5) and (6), the calculated results of the piezoelectric coefficients as function of the thickness fraction k are presented in Fig. 2. Note that the piezoelectric coefficient d_{33} of single crystal BTO (= 165 pm/V) is also shown for comparison. We can know that the effects of the substrate contribution play a significant role in the converse piezoelectric response of a ferroelectric film. The reduction of the piezoelectric coefficient $d^f_{33,\varepsilon}$ from the single crystal value due to substrate clamping, is shown with green arrows. It is consistent with the results reported by Roytburd [17]:

$$d_{33} - d^f_{33,\varepsilon}(k) = \frac{2(1-k)S^f_{13}d_{31}}{k(S^f_{11}+S^f_{12})+(1-k)(S^s_{11}+S^s_{12})}$$

for an epitaxial (001) tetragonal ferroelectric film grown on a (001) cubic substrate. From the above equation, we also know that the reduction diminishes as k reaches 1. On the other hand, the gap between the piezoelectric coefficients $d^f_{33,\varepsilon}$ and $d^f_{33,d}$ is denoted by purple arrows, which represents the contribution of substrate's Poisson strain to $d^f_{33,d}$ and provides partial compensation to the effect of clamping imposed by the substrate. In the Roytburd report [17], this contribution was defined by:

$$d^f_{33,d}(k) - d^f_{33,\varepsilon}(k) = \frac{2(1-k)S^s_{12}d_{31}}{k(S^f_{11}+S^f_{12})+(1-k)(S^s_{11}+S^s_{12})}$$

TABLE I

The elastic coefficients for constant field of Si, $SrTiO_3$, $LaAlO_3$ and MgO at room temperature. It has $S_{11} = S_{22} = S_{33}$, $S_{44} = S_{55} = S_{66}$, $S_{12} = S_{21} = S_{13} = S_{31} = S_{23} = S_{32}$ for the cubic phase, and $S_{11} = S_{22}$, $S_{44} = S_{55}$, $S_{12} = S_{21}$, $S_{13} = S_{31} = S_{23} = S_{32}$ for tetragonal phase.

Constants	S^E_{11}	S^E_{33}	S^E_{12}	S^E_{13}	S^E_{44}	S_{66}
(unit)	$[10^{-12}\ m^2\ N^{-1}]$					
$BaTiO_3$	11.2	23.2	–1.3	–8.2	54	8.1
Si	7.74		–2.17		12.6	
$SrTiO_3$	3.77		–0.93		8.23	
$LaAlO_3$	4.31		–1.27		6.49	
MgO	4.00		–0.98		6.41	

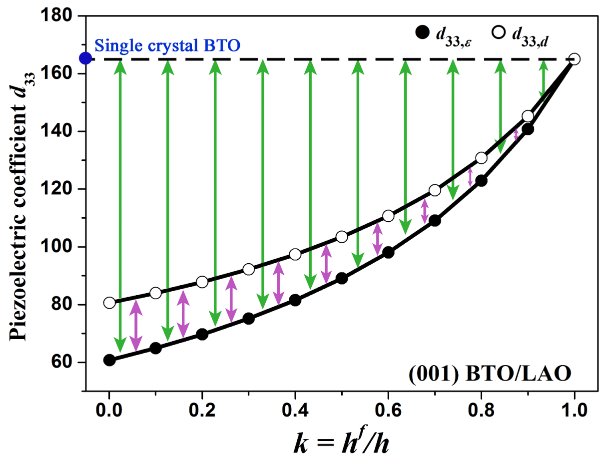

Fig. 2. Comparison of piezoelectric coefficients of single crystal BTO, epitaxial BTO film on (001) LAO substrate and BTO/LAO bilayer heterostructure.

in $(001)_T/(001)_C$ epitaxial heterostructure. It can be see from Fig. 2, there is a tendency for a decrease in the gap between the coefficients $d^f_{33,\varepsilon}$ and $d^f_{33,d}$, and approaches zero as k approaches 1 (the same as a free standing film). $d^f_{33,d}(k) - d^f_{33,\varepsilon}(k)$ is always positive in the range of $0 \leq k < 1$, i.e., the piezoelectric coefficient $d^f_{33,d}$ is always larger than $d^f_{33,\varepsilon}$ and it approaches $d^f_{33,\varepsilon}$ if the substrate is absolutely rigid ($S^s_{12} = 0$) or all substrate boundaries (around sides and underside) are fixed.

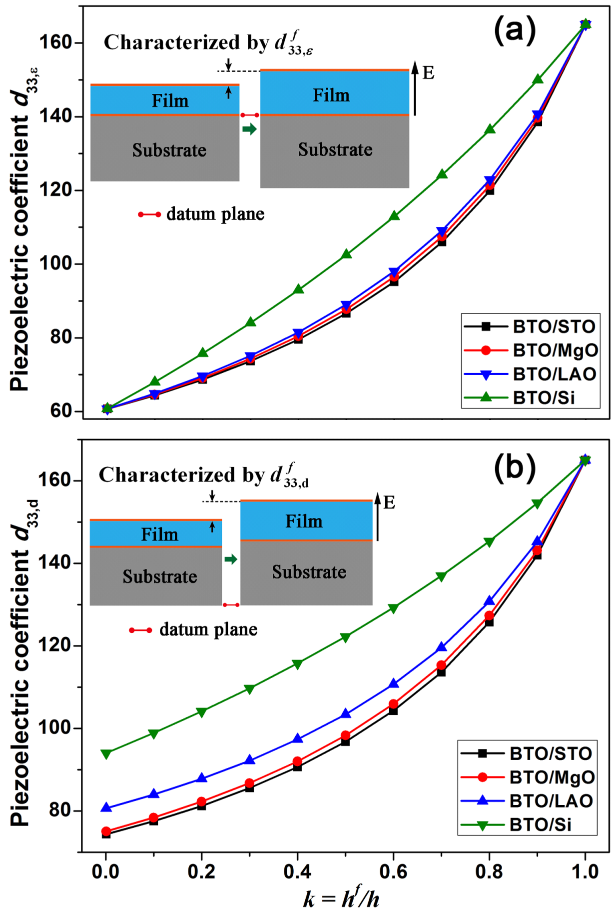

Fig. 3. The piezoelectric coefficients for tetragonal BTO film as functions of the thickness fraction k on (100) Si, STO, LAO and MgO substrates: (a) for BTO film only, (b) for the film/substrate bilayer heterostructure. The insets in (a) and (b) are schematics of the corresponding piezoelectric coefficients, respectively.

In order to illustrate the influence of substrate elastic properties on the piezoelectric performance of ferroelectric films, the computed results of (001) BTO epitaxial films grown on Si, STO, LAO and MgO substrates are presented in Fig. 3. As shown in Fig. 3a, it has the largest value of $d^f_{33,\varepsilon}(k)$ for BTO/Si heterostructure and the smallest for BTO/STO for any given fraction $0 < k < 1$. This indicates that Si is most soft in these four kinds of substrates, $\overline{S}(k)_{(\mathrm{BTO/Si})} > \overline{S}(k)_{(\mathrm{BTO/LAO})} > \overline{S}(k)_{(\mathrm{BTO/MgO})} > \overline{S}(k)_{(\mathrm{BTO/STO})}$, where $\overline{S}(k) = k(S^f_{11} + S^f_{12}) + (1-k)(S^s_{11} + S^s_{12})$ is the average planar elastic compliance of the couple with a $(001)_T/(100)_C$ heteroepitaxy. On the other hand, in Fig. 3b, we can see that it always has a largest piezoelectric coefficient $d^f_{33,d}$ for $\mathrm{BTO}_{(001)}/\mathrm{Si}_{(001)}$ hetero-epitaxial-structure in the range of $0 < k < 1$. A larger $d^f_{33,\varepsilon}(k)$ piezoelectric coefficient on Si substrate than on others can be easily explained by a less effective substrate clamping. On the other hand, because of the contribution of in-plane stresses on the surface piezo-displacement of the heterostructure, the effect of constraint on film will be compensated partially by the elastic deformation of the substrate [16, 17]. On top of the reduced clamping effect, the softest Si substrate provides the highest contribution to the Poisson strain, leading to the largest $d^f_{33,d}(k)$. The clamping effect and substrate deformation should be considered comprehensively in evaluating the piezoelectric performance due to that the changes of them are not synchronized with k. Therefore, for a ferroelectric film epitaxially grown on four kinds of different substrates in present case, the d^f_{33} piezoelectric coefficients, determined by the characterization method ("only film strain" or "total surface displacement") and film thickness fraction, can be about the same or very much different. The corresponding schematics of two piezoelectric coefficients $d^f_{33,\varepsilon}$ and $d^f_{33,d}$ are shown in the inset in Fig. 3a,b, respectively.

It also can be seen in Fig. 3a, the $d^f_{33,\varepsilon}(k)$ piezoelectric coefficient always increases with increasing film thickness fraction k due to reduced clamping effect of substrate. On the other hand, it should be noted that the $d^f_{33,d}$ piezoelectric coefficient can be larger than the intrinsic d_{33} if the substrate is softer than the film in the plane [16, 17, 21], i.e., $|S^s_{12}| > \left|S^f_{13}\right|$. It is not the case for epitaxial BTO film grown on present these substrates, which are harder than BTO film. From the above results in our case as are shown in Fig. 2 and Fig. 3, we can found that, for the (001) epitaxial heterostructure, the $d^f_{33,\varepsilon}$ and $d^f_{33,d}$ coefficients increase accelerative with film thickness. This is characteristic of a soft-film grown on a hard-substrate in terms of the in-plane elasticity.

4. Conclusion

We have studied the piezoelectric properties of a ferroelectric film–substrate elastic bilayer epitaxial heterostructure in this work. Two kinds of longitudinal piezoelectric coefficients $d^f_{33,\varepsilon}$ and $d^f_{33,d}$ were quantitatively solved, and the influence of the fraction of film in bilayer, and the elastic properties of the substrate (different substrate materials) were considered. The results show that the substrate deformation and clamping effect play an important role in evaluating longitudinal piezoelectric responses of a ferroelectric film epitaxial grown on a supporting substrate. All the results presented above are only based on the electromechanical analysis, therefore they are also valid for non-ferroelectric piezoelectric films.

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