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Influence of interface intermixing and periodicity on internal electric field and polarization in ferroelectric superlattices

Khian-Hooi Chew[a,*], Kok-Geng Lim[a], Lye-Hock Ong[b], Makoto Iwata[c]

[a]Department of Physics, University of Malaya, 50603 Kuala Lumpur, Malaysia

[b]School of Physics, Universiti Sains Malaysia, 11800 Minden, Penang, Malaysia

[c]Department of Engineering Physics, Electronics and Mechanics, Graduate School of Engineering, Nagoya Institute of Technology, Nagoya 466-8555, Japan

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Abstract

A thermodynamic model is developed to study the effect of interface intermixing and modulation period on the internal electric field of a superlattice comprising alternate layers of ferroelectric and dielectric. The intermixed layer plays an important role in determining both the polarization and internal electric field modulation profiles of the superlattice. The presence of interface intermixed layer gives rise to inhomogeneous internal field and polarization, which enhances the depolarization field of the superlattices. Interface intermixing; however, does not have much influence on the polarization behavior of the superlattice. Ferroelectric volume fraction or thickness ratio of ferroelectric to dielectric is one of the key parameter for controlling the properties of a superlattice consisting alternate layers of ferroelectric and dielectric.

Keywords: B. Interfaces; C. Ferroelectric properties; Superlattices

1. Introduction

The studies of ferroelectric superlattices have attracted immense attention due to its fundamental scientific interest and potential applications. In layered structures, intermixed layers may form at interfaces between two layers. Intermixed layers can be formed by short-range interactions between two materials, surface reconstruction, cation intermixing or composition deviations at the interfaces in superlattices of ferroelectric solid solution [1].

Using the chemical solution deposition method, Hosokura and co-workers demonstrated the fabrication of $BaTiO_3$/$SrTiO_3$ superlattices without intermixing at interfaces and with good stoichiometry of the deposited films [2]. However, intermixing at interfaces in layered ferroelectrics is usually difficult to control experimentally at high temperature using high-energy lasers, where the stoichiometry of the deposited films changes in a complicated manner under the prescribed deposition conditions [3,4]. Those factors usually promote the formation of intermixed layer at the interface. These intermixed layers with properties different from its constituent layers may affect the properties of the structure [1,3,5].

A detailed evaluation of interface diffusion or intermixing in $BaTiO_3$/$SrTiO_3$ superlattices grown by molecular beam epitaxy was performed by Ishibashi and co-workers [6]. Hung et al. [7] identified the presence of compositional intermixing at interfaces in $PbZrO_3$/$BaZrO_3$ superlattices. A recent study on the structural evolution of surfaces during the layer-by-layer growth of $BaTiO_3$ films on $SrRuO_3$ indicates that the surface reconstruction of $SrRuO_3$ increases the oxygen concentration, and leads to both intermixing and structural change in $BaTiO_3$ at the interface [8]. Mizoguichi et al. [3] carried out a detail study on interface intermixing in $SrTiO_3$-based superlattices, and developed a method to control the intermixing and improve the superlattice properties.

While many thermodynamic models have been put forward to study ferroelectric superlattices [9–15], they all generally do not take into account of intermixing at interfaces. Pertsev and Tyunina [1] proposed a thermodynamic model to study the dielectric permittivity of superlattice by introducing an interface layer with properties different from those of both layers. In their model; however, the effect of interface is not considered. The polarization is homogenous

*Corresponding author. Tel.: +603 7967 4022; fax: +603 7967 4146.
E-mail address: khchew@um.edu.my (K.-H. Chew).

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throughout the layers and the local polarization coupling at interface is neglected. A recent study of local structural distortions in $PbTiO_3/SrTiO_3$ superlattices [16,17] demonstrated the presence of inhomogeneous ferroelectric properties in superlattices. Their works indicated that theoretical studies [1,10,11,13,14] based on the assumption of uniform polarization throughout the layers in superlattices most likely not valid.

We have proposed a thermodynamic model to study the ferroelectric properties of a superlattice [18–20]. An interface energy term is introduced to describe the local polarization coupling at interfaces. The existence of interface polarization coupling leads to the formation of intermixed layer at interfaces [21] and a periodic modulation in polarization [19,20]. We further extended the model [18–20] to include the internal electric field and epitaxial strain, enabling it to compare with experimental data [22].

In this paper, we aim to investigate the effects of interface intermixing and modulation period on the internal electric field and polarization of a superlattice comprising alternate layers of ferroelectric and dielectric. The correlation between the internal electric field and ferroelectric properties of the superlattice is examined by looking at the modulation profiles. The effect of interface intermixing and modulation period on the internal electric field and polarization are studied by changing the volume fraction or thickness ratio of the superlattice.

2. Theory

We consider a superlattice composed of two constituents: a ferroelectric layer and a dielectric layer (hereafter, we denote the superlattice as a ferroelectric/dielectric superlattice), which grows on a substrate, as schematically shown in Fig. 1.

By assuming that all spatial variation of polarization takes place along the z-direction, the Helmholtz free energy [23] per unit area for one period of the superlattice can be expressed as the following:

$$F = \int_{-L_f}^{0} f_f dz + \int_0^{L_d} f_d dz + f_i \tag{1}$$

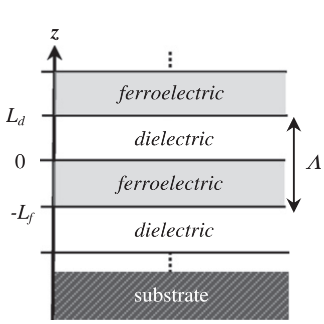

Fig. 1. Schematic illustration of a periodic superlattice composed of a ferroelectric f layer and a dielectric d layer. The thicknesses of f and d layers are L_f and L_d, respectively. Λ denotes the modulation period of the ferroelectric/dielectric superlattice.

where the first and second terms denote the free energy per unit area of ferroelectric (f) layer with thickness L_f and the free energy per unit area of dielectric (d) layer with thickness L_d, respectively. In Eq. (1), f_j(j:f or d) denotes the free energy density as follows [23]:

$$f_j = \alpha_j^* p_j^2 + \beta_j^* p_j^4 + \gamma_j p_j^6 + \frac{\kappa_j}{2}\left(\frac{dp_j}{dz}\right)^2 + \left(\frac{c_{11,j}^2 + c_{11,j}c_{12,j} - 2c_{12,j}^2}{c_{11,j}}\right) u_{m,j}^2 - \frac{1}{2} e_{dep,j} p_j \tag{2}$$

where p_j represents the polarization of j layer. $\alpha_j^* = \alpha_j + 2\left(c_{12j}g_{11j}/c_{11j} - g_{12j}\right)u_{mj}$ and $\beta_j^* = \beta_j - g_{11j}^2/2c_{11j}$ where α_j (temperature dependent), β_j and γ_j are the Landau coefficients. c_{11j} and c_{12j} are the elastic stiffness coefficients, whereas g_{11j} and g_{12j} corresponds to the electrostrictive constants. $u_{mj} = (a_S - a_j)/a_S$ denotes the in-plane misfit strain induced by the substrate due to the lattice mismatch. a_j is the unconstrained equivalent cubic cell lattice constants of j layer, whereas a_S is the lattice parameter of the substrate. κ_j is the gradient coefficient that determines the energy cost due to the inhomogeneity of polarization p_j within the layer, $e_{dep,j}$ corresponds to the internal electric field of j layer.

The interface energy f_j in Eq. (1) is [18–22]:

$$f_i = \frac{\lambda_0}{2\varepsilon_0}\left[(p_f(0) - p_d(0))^2 + (p_f(-L_f) - p_d(L_d))^2\right] \tag{3}$$

where λ_0 is the temperature-independent interface parameter and λ_0 is the dielectric permittivity in vacuum. By symmetry, the polarization (or induced-polarization) at interfaces of the ferroelectric and dielectric layers can be expressed as $p_f(0) = p_f(-L_f) = p_{fi}$ and $p_d(0) = p_d(L_d) = p_{di}$, respectively. Hence, Eq. (3) can be reduced to $f_i = \frac{\lambda_0}{\varepsilon_0}(p_{fi} - p_{di})^2 = \frac{\lambda_0}{\varepsilon_0}(p_{fi}^2 + p_{di}^2) - \frac{2\lambda_0}{\varepsilon_0} p_{fi} p_{di}$. This implies that the physical origin of the interface energy term can be unambiguously understood by separating the energy term into two components [22]: (i) non-ferroelectric component $\lambda_0(p_{fi}^2 + p_{di}^2)/\varepsilon_0$ and (ii) polarizations coupling component $2\lambda_0 p_{fi} p_{di}/\varepsilon_0$. The former component is analogous to the formation of "dead" layers [24] at interfaces. The latter component describes the coupling between the local polarizations at interfaces due to the modification of bonding at the interfaces. In the present model, λ_0 describes the intermixing at interface in superlattices. If $\lambda_0 \neq 0$, intermixed layers [21] (analogous to a dead layer [24]) with properties different from those of both constituents is expected to form at the interface region. No intermixed layer is formed, if $\lambda_0 = 0$.

The internal electric field $e_{dep,j}$ of layer j can be obtained by the Maxwell's equations that are, in the present case with no free charges [15,22]

$$\nabla(\boldsymbol{\varepsilon_0}\mathbf{E} + \mathrm{P}) = 0 \tag{4}$$

$$\nabla \times \mathbf{E} = 0 \tag{5}$$

where $\mathbf{E}$ and $\mathbf{P}$ denote the electric field and polarization, respectively. Eq. (5) implies that we can define a scalar function φ_j, i.e. the electrostatic potential, which satisfies $e_{dep,j} = -\nabla\varphi_j$. The Euler-Lagrange equations can be expressed in term of ϕ_j using $e_{dep,j} = -\nabla\varphi_j$ as follows [22]:

$$\kappa_j \frac{d^2 p_j}{dz^2} = 2\alpha_j^* p_j + 4\beta_j^* p_j^3 + 6\gamma_j p_j^5 + \frac{1}{2}\frac{d\varphi_j}{dz} \tag{6}$$

with the electrostatic potentials

$$\varepsilon_0 \frac{d^2\varphi_j}{dz^2} + \frac{dp_j}{dz} = 0 \tag{7}$$

which can be found from the Maxwell's Eq. (4). At the interface, the boundary conditions for the polarization are

$$\begin{cases} -\kappa_f \left.\dfrac{dp}{dz}\right|_{z=-L_f} + \dfrac{\lambda_0}{\varepsilon_0}\left[p_f(-L_f) - p_d(L_d)\right] = 0 \\ \kappa_d \left.\dfrac{dp}{dz}\right|_{z=0} + \dfrac{\lambda_0}{\varepsilon_0}\left[p_f(0) - p_d(0)\right] = 0 \\ \kappa_f \left.\dfrac{dp}{dz}\right|_{z=0} + \dfrac{\lambda_0}{\varepsilon_0}\left[p_f(0) - p_d(0)\right] = 0 \\ -\kappa_d \left.\dfrac{dp}{dz}\right|_{z=L_f} + \dfrac{\lambda_0}{\varepsilon_0}\left[p_f(-L_f) - p_d(L_d)\right] = 0 \end{cases} \tag{8}$$

and the electrostatic boundary conditions are

$$\begin{cases} -\varepsilon_0 \left.\dfrac{d\varphi_f}{dz}\right|_{z=0} + \varepsilon_0 \left.\dfrac{d\varphi_d}{dz}\right|_{z=0} = -(p_f(0) - p_d(0)), \\ -\varepsilon_0 \left.\dfrac{d\varphi_f}{dz}\right|_{z=-L_f} + \varepsilon_0 \left.\dfrac{d\varphi_d}{dz}\right|_{z=L_d} = -(p_f(-L_f) - p_d(L_d)), \end{cases} \tag{9}$$

with the potentials

$$\begin{cases} \varphi_f(0) = \varphi_d(0), \\ \varphi_d(-L_f) = \varphi_d(L_f). \end{cases} \tag{10}$$

In this work, the Eqs. (6) and (7) were solved using the finite-difference method subject to the boundary conditions of Eqs. (8) to (10).

3. Results and discussion

As an illustration, we apply the model to a superlattice consisting of $PbTiO_3$(PT) as ferroelectric layers and $SrTiO_3$(ST) as dielectric layers, grown on ST substrate. The thermodynamic parameters of bulk PT and bulk ST used in the calculation are listed in Table 1. In the calculations, we take 1 unit cell (u.c.) as ~0.4 nm [25]. $\xi_0 = \sqrt{\kappa_f/(\alpha_{0f}T_{0f})} \sim 0.6$ nm is defined as the characteristic length of domain wall half width [12,25]. At $T=298K$, the calculated bulk polarization of PT is $P_0 \sim 0.75$ C/m^2. The lattice constants in the paraelectric state are $a_f = 3.969$ Å and $a_d = 3.905$ Å for PT and ST, respectively [13]. Based on the lattice constants, the misfit strains with PT and ST (due to the ST substrates) are $u_{mf} = -0.0164$ and $u_{md} = 0$, respectively.

Fig. 2 shows the spatial dependence of internal electric field and polarization of PT/ST superlattices with $L_f =$ $L_d = 4$ *u.c.* for different λ_0 values. If $\lambda_0 = 0$, no intermixing is expected. Both the internal electric field and polarization remains homogeneous throughout the ferroelectric or dielectric layers. For the case of superlattices with $\lambda_0 \neq 0$, an intermixed layer, with properties different from those of both layers, is formed at interface $z=0$ [21]. The formation of intermixed layer at interfaces leads to inhomogeneous polarization and internal electric field near the interfaces. The continuity or discontinuity of polarization and internal electric field across the interface depends sensitively on the nature of intermixed layer. The intermixed layer plays an important role in determining the polarization [19,20] and internal electric field [22] modulation profiles of internal electric field and polarization in the superlattice. In addition, both the magnitude and gap of polarization and internal electric field decreases with increasing λ_0 value.

From Fig. 2, it is seen that the internal electric field in PT layer acts as the depolarization field $e_{dep,f} < 0$. On the

Table 1
List of thermodynamic parameters used in this study [13].

	PT	ST	Units
α	$3.8\ (T - T_{0PT})$	$7.45\ (T - T_{0ST})$	$\times 10^5$ JmC^{-2}
β	4.229	2.02	$\times 10^8$ Jm^{5}C^{-4}
γ	2.6	–	$\times 10^8$ Jm^{9}C^{-6}
T_0	752	51.64	K
g_{11}	1.14	1.25	$\times 10^{10}$ JmC^{-2}
g_{12}	4.63	−10.8	$\times 10^8$ JmC^{-2}
c_{11}	1.746	3.36	$\times 10^{11}$ Jm^{-3}
c_{12}	0.794	1.07	$\times 10^{11}$ Jm^{-3}
κ	1.029	1.029	$\times 10^{-10}$ Jm^{3}C^{-2}

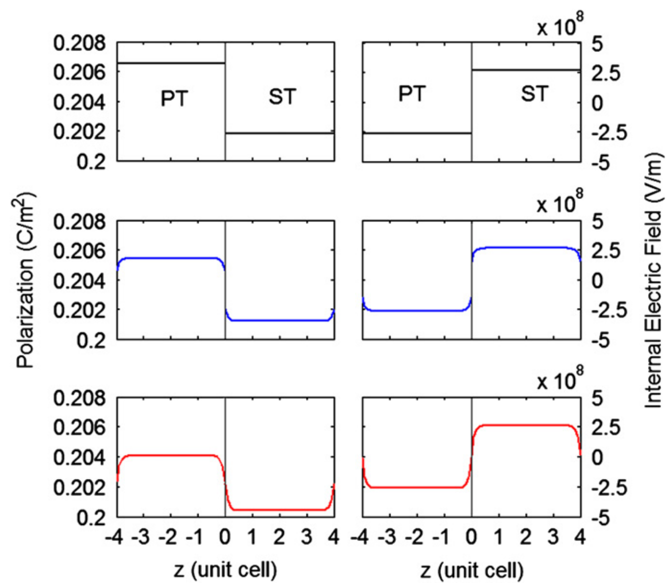

Fig. 2. Profiles of internal electric field and polarization of PT/ST superlattices with $L_f = L_d = 4$ u.c. at $T = 298$ K. The values of λ_0 are: 0(black), 0.02 ξ_0 (blue) and ξ_0 (red). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

other hand, $e_{dep,d}>0$ induce the polarization in ST layer. Hereafter, we denote it as "polarization-induced internal electric field". The internal electric field in ST layer originates from the electrostatic interaction between polarizations in different PT layers across the ST layer [22]. Note also that the internal field-induced polarization in the ST layer is almost the same as the spontaneous polarization of the PT layer, implying that the "polarization-induced internal electric field" in ST layer plays an important role in governing the polarization behavior of a superlattice.

We now examine the periodicity dependence of internal electric field and polarization in PT/ST superlattices. In Fig. 3, we show the internal electric field and polarization as a function of period thickness for a PT/ST superlattice with ferroelectric volume fraction $\phi = L_f/(L_f+L_d)=0.5$ (or thickness ratio of ferroelectric to dielectric $L_f/L_d=0.5$). The average polarization is defined as $P=1/\Lambda(\int_{-L_f}^{0} f_f dz+\int_0^{L_d} f_d dz)$ with the periodic thickness Λ. Similarly, the average internal electric fields is defined as $e_{dep}=1/\Lambda(\int_{-L_f}^{0} e_{dep,f} dz+\int_0^{L_d} e_{dep,d} dz)$. Without intermixing (black), both internal field and polarization are essentially independent from changing the layer thickness $L_f=L_d=L$. The depolarization field in PT layer and polarization-induced internal electric field in ST layer completely compensate each other, resulting in a zero net internal field in the superlattice. While the formation of intermixed layer gives rise to the existence of depolarization field, it has little influence on the polarization behavior of the PT/ST superlattices (blue and red). The enhancement of depolarization field is clearly due to the inhomogeneous polarization at the interface region.

The internal electric field and polarization as a function of period thickness for a particular case of PT/ST superlattices with $\lambda_0=\xi_0$ are illustrated in Fig. 4, where the feroelectric volume fraction ϕ is varied from 0.33 to 0.96

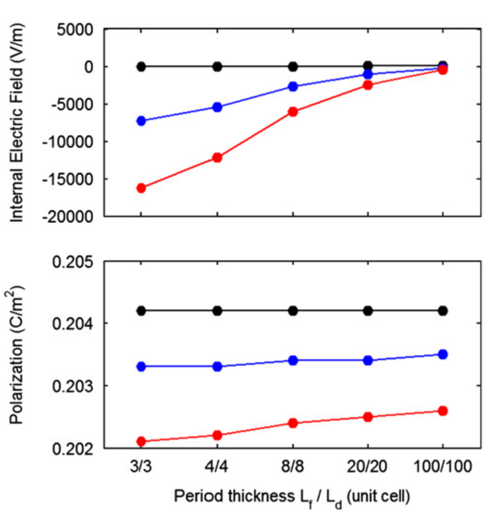

Fig. 3. Internal electric field and polarization as a function of period thickness L_f/L_d for PT/ST superlattices with $L_f=L_d$ at $T=298$ K. The values of $\lambda_0$are: 0 (black), 0.02 ξ_0 (blue) and ξ_0 (red). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

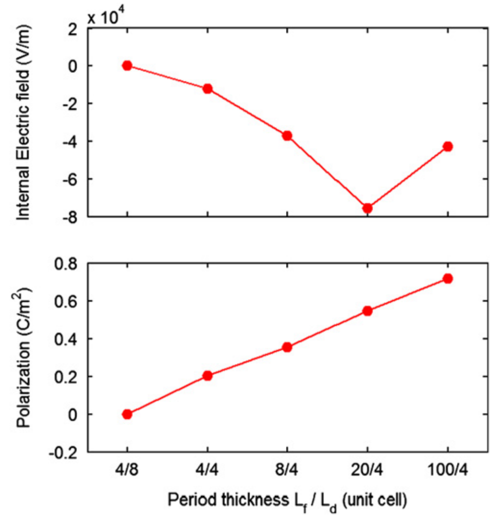

Fig. 4. Internal electric field and polarization as a function of period thickness L_f/L_d for PT/ST superlattices with $L_f \neq L_d$ and $\lambda_0=\xi_0$ at $T=298$ K. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

(or the thickness ratio L_f/L_d is changed from 0.2 to 25). It is seen that the polarization increases as ϕ increases. As ϕ increases from 0.33 to 0.83, the magnitude of depolarization field increases. Upon further increasing ϕ from 0.83 to 0.96, the magnitude of depolarization field reduces. This is because PT approaches its bulk, and intermixing at interface no longer has a strong effect on the internal electric field.

From Figs. 3 and 4, the properties of superlattice remain almost unchanged as the ferroelectric volume fraction ϕ or thickness ratio L_f/L_d is fixed irrespective of varying the layer thickness (as shown in Fig. 3).On the other hand, changing the volume fraction ϕ from 0.33 to 0.96 (or the thickness ratio L_f/L_d from 0.33 to 25) allows the tuning of polarization from ~0 to 0.72 C/m^2. For the superlattice with period thickness 100/4 (Fig. 4), the polarization is found to be ~0.72 C/m^2, which is almost 4 times larger that of the 100/100 superlattice (Fig. 3). Ferroelectric volume fraction ϕ or thickness ratio L_f/L_d, therefore, is one of the key parameter that allows the tuning of ferroelectric properties in superlattice consisting of ferroelectric and dielectric layers [13].

4. Conclusion

We have studied the effect of interface intermixing and periodicity on internal electric field and polarization in superlattices consisting of ferroelectric and dielectric layers. Our results indicate that intermixing at interface gives rise to an inhomogeneous polarization, and enhances the depolarization field of the superlattices. The existence of intermixing at interface plays an important role in determining the polarization and internal field modulated profiles. However, interface intermixing does not have a significant effect on the polarization behavior of the superlattice. Ferroelectric volume fraction $L_f/(L_d+L_f)$ or thickness ratio L_f/L_d is one of the key parameters that controls the properties in a superlattice consisting of ferroelectric and dielectric layers.

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