

Polarization switching of the incommensurate phases induced by flexoelectric coupling in ferroelectric thin films

Limei Jiang,^{a,b} Yichun Zhou,^{a,b,*} Yi Zhang,^a Qiong Yang,^a Yijia Gu^c and Long-Qing Chen^c

^aHunan Provincial Key Laboratory of Thin Film Materials and Devices, School of Materials Science and Engineering, Xiangtan University, Xiangtan 411105, Hunan, China

^bKey Laboratory of Key Film Materials & Application for Equipment of Hunan province, Xiangtan University, Xiangtan 411105, Hunan, China

^cDepartment of Materials Science and Engineering, The Pennsylvania State University, University Park, PA 16802, USA

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Abstract—The polarization switching of the incommensurate (INC) phases induced by flexocoupling in perovskite ferroelectric thin films is investigated with a multi-field coupling theoretical framework combining the flexoelectric effect. The dominant factors of the formation of INC phases that show antiferroelectric-like double hysteresis loops are examined. The simulations show that mechanical boundary conditions have little influence on the polarization responses of INC phases. The polarization switching behaviors of INC phases are governed by the flexocoupling types described by different flexocoupling coefficients. Only the transverse flexocoupling coefficient related INC phases show antiferroelectric-like double hysteresis loop. The longitudinal flexocoupling coefficient related and shear flexocoupling coefficient related INC phases show imprint-like hysteresis loops and hysteresis loops similar to those of the ferroelectric phase, respectively. The observed different polarization switching behaviors are rationalized by free energy density curves of the INC phases.

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1. Introduction

In the past four decades, the INC phases have been observed in a large number of ferroelectric materials, and have attracted considerable interests of scientists [1–6]. The INC phase is defined as the phase in which the lattice period has a sinusoidal spatial modulation [2,3]. Many efforts have been taken to explore the driving forces that are responsible for the appearance of the INC phase [7–10]. One of the most likely driving forces is flexoelectric coupling [11–15], which describes the generation of an electric field by a strain gradient (the direct flexoelectric effect) or the mechanical response induced by an electric field gradient (the converse flexoelectric effect):

$$E_i = -f_{klj} \frac{\partial \varepsilon_{kl}}{\partial x_j} \quad (1a)$$

$$\sigma_{ij} = f_{ijkl} \frac{\partial P_k}{\partial x_l} \quad (1b)$$

* Corresponding author at: Hunan Provincial Key Laboratory of Thin Film Materials and Devices, School of Materials Science and Engineering, Xiangtan University, Xiangtan 411105, Hunan, China. Tel.: +86 731 58293586; fax: +86 731 58292468.; e-mail: zhouyc@xtu.edu.cn

Here, E_i , σ_{ij} , ε_{kl} , P_k and x_j are electric field, stress, strain, polarization, and position coordinate, respectively. f_{ijkl} are components of the flexoelectric tensor. Already in 1970, Axe et al. [1] pointed out that the formation of a modulated INC phase in potassium tantalate is a result of flexoelectric coupling. Since then, many scientists including Blinc and Levanyuk [16], Petzelt [4], Maclaren et al. [5] and Ahluwalia et al. [6] confirmed this point of view in the study of the INC phase in ferroelectric materials.

As we know, macro electric properties of ferroelectrics are closely related to the polarization switching [15,17–19]. Since polarization distribution in an INC phase is totally different from that in a ferroelectric phase [6], it is natural to expect that the polarization switching in an INC phase is also dissimilar to that in a ferroelectric phase. Therefore, ferroelectrics in the flexoelectric coupling related INC phases are predicted to have intriguing macro electric properties. In other words, the INC phase may be the physic origin of some abnormal properties of ferroelectric materials. In fact, Tagantsev et al. [10] revealed that the antiferroelectric state in lead zirconate is a ‘missed’ INC phase, and that the paraelectric to antiferroelectric phase transition is driven by the softening of a single lattice mode via flexoelectric coupling. Our previous study [20] demonstrated that the INC phase induced by the f_{1122} related coupling shows antiferroelectric-like double hysteresis loops

under a high electric field. However, though we have realized that the INC phases induced by flexocoupling can present intriguing macro electric properties, our understanding of the INC phases is still rather incomplete. There are still no studies investigating the dominant factor in the development of INC phases that show antiferroelectric-like properties. Also, there are still no studies clarifying whether flexoelectric coupling related INC phases can show other unexpected properties.

To address these issues, a multi-field coupling theoretical framework of flexoelectric effect, which is developed in our previous work [20], is adopted to study the polarization switching of the flexocoupling-driven INC phases in perovskite ferroelectric thin films. In order to clarify whether the mechanical boundary condition is a key factor in development of INC phases that show antiferroelectric-like property, the mechanical boundary conditions in the present study are different from those in our previous study. The ferroelectric thin film here are constrained by two electrodes, while in our previous study only the bottom surface of the ferroelectric thin film is constrained. For a material of cubic point group, there are three independent components of the flexoelectric coefficients: f_{1111} , f_{1122} , and f_{1212} ($=f_{2121}$), describing three different flexocoupling types. For the purpose of examining whether the flexocoupling type is a dominant factor in formation of INC phases that show antiferroelectric-like property and clarifying whether INC phases can show other unexpected properties, we study the polarization switching of INC phases induced by three different flexocoupling types: (a) $f_{11} \neq 0$, $f_{12} = f_{44} = 0$, (b) $f_{12} \neq 0$, $f_{11} = f_{44} = 0$, (c) $f_{44} \neq 0$, $f_{11} = f_{12} = 0$, where f_{11} , f_{12} , and f_{44} are respectively the Voigt form of the tensors f_{1111} , f_{1122} , and f_{1212} .

The simulation results predict that the mechanical boundary condition is not a key factor that governs the polarization response of the INC phases though it can affect the polarization distributions of the INC phases and the threshold value at which the INC phase formed. The polarization switching is mainly influenced by the flexoelectric coupling type. Only the f_{12} coupling related INC phases show antiferroelectric-like double hysteresis loops. The other two flexocoupling type induced INC phases demonstrate distinct polarization switching behaviors.

2. Theoretical framework for electromechanical coupling behavior considering flexoelectric effect

Three sets of equations should be included to establish the multi-field coupling theoretical framework of flexoelectricity, they are [20]

(a) Thermodynamic potential considering flexoelectric effect

Generally, it requires three physical fields, namely the polarization field, electric field and displacement field, to describe the multi-field coupling behavior of ferroelectric materials under electrical and mechanical loadings [21]. The quantities utilized to depict these three physical fields are displacement (\mathbf{u}), stress ($\boldsymbol{\sigma}$), strain ($\boldsymbol{\varepsilon}$), double stress (\boldsymbol{t}), strain gradient ($\nabla\boldsymbol{\varepsilon}$), electric potential (φ), electric field (\mathbf{E}), electric displacement (\mathbf{D}), polarization (\mathbf{P}) and polarization gradient ($\nabla\mathbf{P}$). Among them, $\boldsymbol{\varepsilon}$, \mathbf{E} , \mathbf{P} , $\nabla\mathbf{P}$ and $\nabla\boldsymbol{\varepsilon}$ are chosen as independent variables [21,22] for the

following numerical implementations. The corresponding thermodynamic potential density for this set of independent variables is electric enthalpy, which can be expressed as

$$h = \frac{1}{2}\alpha_{ij}P_iP_j + \frac{1}{4}\beta_{ijkl}P_iP_jP_kP_l + \frac{1}{6}\gamma_{ijklmn}P_iP_jP_kP_lP_mP_n + \frac{1}{2}c_{ijkl}\varepsilon_{ij}\varepsilon_{kl} - \frac{1}{2}q_{ijkl}\varepsilon_{ij}P_kP_l + \frac{1}{2}g_{ijkl}P_{i,j}P_{k,l} - \frac{1}{2}\varepsilon_0k_{ii}E_iE_i - E_iP_i + \frac{1}{2}f_{ijkl}\left(\frac{\partial P_i}{\partial x_j}\varepsilon_{kl} - P_k\frac{\partial\varepsilon_{ij}}{\partial x_l}\right) \quad (2)$$

where the indices run from 1 to 3, repeating indices imply summation, and commas imply differentiation. The first three terms in Eq. (2) represent the Landau free energy density, where α_{ij} , β_{ijkl} , γ_{ijklmn} are the phenomenological Landau-Devonshire coefficients [23]. The fourth term denotes the elastic energy density of the system, in which c_{ijkl} are the elastic constants. The fifth term denotes the coupling energy density between the polarizations and the strains, where q_{ijkl} are the electrostrictive coefficients. The sixth term is the polarization gradient energy density, in which g_{ijkl} are the gradient energy coefficients. The seventh and eighth terms denote the electric energy density due to the presence of an electric field, where ε_0 is the dielectric permittivity of vacuum and k_{ii} are the background dielectric constants of the ferroelectric film. The last term denotes the flexoelectric coupling energy density, where f_{ijkl} are components of the flexocoupling tensor.

(b) Constitutive equations of each physical field with flexoelectricity

The constitutive equations of each physical field can be derived from the electric enthalpy density defined in Eq. (2), i.e.

$$t_{ijl} = \frac{\partial h}{\partial\varepsilon_{ij,l}}, \sigma_{ij} = \frac{\partial h}{\partial\varepsilon_{ij}}, D_i = \frac{\partial h}{\partial E_i}, \eta_i = \frac{\partial h}{\partial P_i}, \Lambda_{ij} = \frac{\partial h}{\partial P_{i,j}} \quad (3)$$

where η_i is the effective local electric force, Λ_{ij} can be taken as higher order local electric force. t_{ijl} , the double stress tensor, work-conjugate with the strain gradient tensor $\varepsilon_{ij,k} = \partial\varepsilon_{ij}/\partial x_k$.

(c) The balance laws and boundary conditions for each physical field

The mechanical equilibrium equation accounting for contributions from strain gradients is given based on the Mindlin's strain gradient theory [24,25]

$$(\sigma_{ik} - t_{ijk,j})_i + b_k = 0 \quad (4)$$

where b_k are the components of a body force per unit volume.

The electric field is governed by the Maxwell's (or Gauss') equation

$$D_{i,i} - \zeta = 0 \quad (5)$$

where ζ is the volume charge density.

The temporal evolution of the polarization field is governed by the time dependent Ginzburg–Landau equations [22,26–30],

$$\frac{\partial P_i}{\partial t} = -L[\eta_i - \Lambda_{ij,j}] \quad (6)$$

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