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Phase-field simulation of strain-induced ferroelectric domain evolution in hexagonal manganites

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ABSTRACT

The domain structures evolution induced by strain in hexagonal manganites was simulated through using phase-field simulation. The result demonstrated that tensile strain in *x* direction (or compressive strain in *y* direction) and pure shear strain will drive topological ferroelectric vortex into topological stripe domains in *y* direction and *x* direction, respectively. During this ferroelectric domain evolution, the annihilation of topological vortex-antivortex was found in a way that vortices and antivortices are moving in the opposite direction. Meanwhile, the topological charge is conservative during this evolution under the periodic boundary condition.

1. Introduction

Hexagonal (*h*-) manganites are multiferroic material with ferroelectricity and antiferromagnetism coexisting at low temperature, which attracts considerable attentions during last decade. The topological defects have been found when the material is cooling above the Curie temperature [1–6], where a structural trimerization is induced, giving rise to three types of antiphase domains (α, β, γ) with each exhibiting two possible directions of induced ferroelectric polarization $(+,-)$ along the *z* direction [7,8]. Therefore there are six types of antiphase and ferroelectric domains [1,9,10]. The domain structure containing topological defects can be viewed as the vortex and anti-vortex in a self-organized network.

Understanding and controlling topological defects in multiferroic materials is crucial for technological applications of functional materials. Due to the thermal annealing details, surface defects or impurities, different types of topological structure can be found [9,11,12]. Recently, experiment demonstrated that the strain applied in multiferroic hexagonal manganites moves vortices and antivortices in opposite directions and stabilizes a topological stripe domain state [13], which indicates that the coupling between strain and order parameters in hexagonal manganites is critical for the topological domain structure and should be analyzed carefully. We should noted that the vortex to stripe domain transformation happens around the ferroelectric-trimerization transition temperature, T_c, which can reach as high as 1450 °C in $LuMnO_3$ crystal, makes it impossible for a direct observation at such high temperature, thus a theoretical model to simulate this process is necessary. Usually the condensation of vortices through the critical phase transition temperature can be analyzed using Monte-Carlo method with *XY* model [14]. Recently, mean field theory based on Landau free energy is employed [15,16], and phase-field methods have been used to predict the domain evolution and topological vortex distributions [17,18]. However, the simulation of the topological vortex domain affected by electric field, strain, defects *etc.*, are rarely discussed. In this paper, we reported that the vortices can be changed to stripe domain when applied a pure tensile strain or shear strain by using phase field simulation which shows a useful technique to investigate the complex topological domain evolution.

2. Theoretical model

Phase-field simulations are widely used for the temporal evolution of domain patterns in ferroelectric or ferromagnetic materials [19,20]. The trimerization of the *h*-$YMnO_3$ can be described by the magnitude Q and the azimuthal angle Φ . The free energy

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density of the system is given by Ref. [15]:

$$f = \frac{a}{2}Q^2 + \frac{b}{4}Q^4 + \frac{Q^6}{6}(c + c'\cos 6\,\Phi) - gQ^3P\cos 3\,\Phi + \frac{g'}{2}Q^2P^2 + \frac{a_P}{2}P^2 \quad (1)$$
$$\frac{1}{2}\sum_{i=x,y,z}\left[s_Q^i\left(\partial_i Q\partial_i Q + Q^2\partial_i\phi\partial_i\phi\right) + s_P^i\partial_i P\partial_i P\right]$$

where a, b, c, c', g, g', a_P are the coefficients for the Landau free energy function, $s_Q^x, s_Q^y, s_Q^z, s_P^x, s_P^y, s_P^z$ are coefficients for the gradient energy terms of trimerization magnitude and the polarization. The nonlinear interaction term between Q and P makes sure that electric polarization induced in the states with is positive (+), the remaining is negative (−). There are six domain walls, and neighboring energy minima are separated by anti-parallel polarization, the structural domain wall with change larger than $\pi/3$ is unfavorable, due to higher gradient energy. Here we consider a two-dimension model in the xy plane (corresponding to ab plane in experimental demonstration) and transform the three phase order parameters to Q_x, Q_y, P_z which have the relation $Q_x = Q\cos\Phi, Q_y = Q\sin\Phi$. Therefore, we obtain [17]:

$$\begin{aligned} f = {} & \frac{a}{2}\left(Q_x^2 + Q_y^2\right) + \frac{b}{4}\left(Q_x^2 + Q_y^2\right)^2 + \frac{c}{6}\left(Q_x^2 + Q_y^2\right)^3 + \frac{c'}{6}\left(Q_x^6 \right. \\ & \left. - 15Q_x^4Q_y^2 + 15Q_x^2Q_y^4 - Q_y^6\right) - g\left(Q_x^3 - 3Q_xQ_y^2\right)P_z \\ & + \frac{g}{2}\left(Q_x^2 + Q_y^2\right)P_z^2 + \frac{a_p}{2}P_z^2 + \frac{s_Q^x}{2}\left[\left(\frac{\partial Q_x}{\partial x}\right)^2 + \left(\frac{\partial Q_x}{\partial y}\right)^2\right. \\ & \left. + \left(\frac{\partial Q_y}{\partial x}\right)^2 + \left(\frac{\partial Q_y}{\partial y}\right)^2\right] + \frac{s_Q^z}{2}\left[\left(\frac{\partial Q_x}{\partial z}\right)^2 + \left(\frac{\partial Q_y}{\partial z}\right)^2\right] \\ & + \frac{s_P^z}{2}\left(\frac{\partial P_z}{\partial z}\right)^2 + \frac{s_P^x}{2}\left[\left(\frac{\partial P_z}{\partial x}\right)^2 + \left(\frac{\partial P_z}{\partial y}\right)^2\right] - E_zP_z + f_{strain} \end{aligned} \quad (2)$$

where E_z is an external electric field along the z direction. f_{strain} is the energy density from the lowest-order coupling of the inhomogeneous trimerization with strains, which has the form [15]:

$$f_{strain} = -GQ^2\left[(u_{xx} - u_{yy})\partial_x\Phi - 2u_{xy}\partial_y\Phi\right] \quad (3)$$

where u_{xx}, u_{yy}, u_{xy} are tensile strain in x, y directions and shear strain, respectively. G is the coupling parameter. In order to unify the phase-field parameter orders to Q_x, Q_y, P_z , we also transform the angle term in strain energy to Q_x, Q_y. First, according to $Q_x = Q\cos\Phi, Q_y = Q\sin\Phi$ we can get $\tan\Phi = \frac{Q_y}{Q_x}$,

$$\begin{aligned} \frac{\partial\Phi}{\partial x} &= \frac{-Q_y}{Q_x^2 + Q_y^2}\frac{\partial Q_x}{\partial x} + \frac{Q_x}{Q_x^2 + Q_y^2}\frac{\partial Q_y}{\partial x} \\ \frac{\partial\Phi}{\partial y} &= \frac{-Q_y}{Q_x^2 + Q_y^2}\frac{\partial Q_x}{\partial y} + \frac{Q_x}{Q_x^2 + Q_y^2}\frac{\partial Q_y}{\partial y} \end{aligned} \quad (4)$$

Through variation and using (4), we can get:

$$\begin{aligned} \delta F_{strain} = -\int \Big[& G\delta(-(u_{xx} - u_{yy})Q_y)\frac{\partial Q_x}{\partial x} + G\delta((u_{xx} - u_{yy})Q_x)\frac{\partial Q_y}{\partial x} \\ & + G(u_{xx} - u_{yy})\left(Q_x\delta\frac{\partial Q_y}{\partial x} - Q_y\delta\frac{\partial Q_x}{\partial x}\right) \\ & - G\delta(2u_{xy}Q_x)\frac{\partial Q_y}{\partial y} + G\delta(2u_{xy}Q_y)\frac{\partial Q_x}{\partial y} \\ & - 2Gu_{xy}\left(Q_x\delta\frac{\partial Q_y}{\partial y} - Q_y\delta\frac{\partial Q_x}{\partial y}\right)\Big]dV \end{aligned} \quad (5)$$

At last the solving equation is time-dependent Ginzburg-Landau equation [21]:

$$\frac{\partial\eta_p(r,t)}{\partial t} = -L\frac{\delta F}{\delta\eta_p(r,t)} + \xi_{\eta_p}(r,t), \eta_p = Q_x, Q_y, P_z \quad (6)$$

where L is the kinetic coefficient, r is the spatial coordinate, t is simulation time, $\xi_{\eta_p}(r,t)$ is the Langevin noise term.

3. Results and discussion

In this calculation, the simulation of size is $1024\Delta x \times 1024\Delta x \times 1\Delta x$ and the grid spacing is $\Delta x = 0.8$nm, finite difference scheme is employed and the periodic boundary condition is used here. The time step is 0.01 which is sufficient for the iteration convergence, the parameters we used was given by Artyukhin et al. [15] as shown in Table 1, which were obtained from the first-principles calculations on $YMnO_3$. The strain coupling parameter multiplied by strain $G(u_{xx} - u_{yy})$ is assumed as 10.0 eVÅ^{-2} which make the strain coupling energy large enough to drive the topological vortex domain motion in the simulation. Here the strain applied is assumed to be homogeneous, and two different cases are considered. The first one is tensile strain along x direction and compress strain along y direction. The second case is pure shear strain.

We evolve the domain structure under zero applied strain with the initial random distribution, which stabilizes the vortex domain. Fig. 1 show the transformation from vortex domain to stripe domain. Different colors in Fig. 1(a) represent different structures order phases Φ, there are six phases with $\Phi = 0, 2\pi/3$ and $-2\pi/3$ (the α_+, β_+ and γ_+ phases) and $\Phi = +\pi/3, \pi$ and $-\pi/3$(the γ_-, α_- and β_- phases). Here the γ_+ phase is the same as the angle of $4\pi/3$, so the cyan color represents the stable structure order phase $-2\pi/3$, the vortex domain is in the order $(\alpha_+, \gamma_-, \beta_+, \alpha_-, \gamma_+, \beta_-)$ in the counter-clockwise direction and the anti-vortex is in the order $(\alpha_+, \gamma_-, \beta_+, \alpha_-, \gamma_+, \beta_-)$ in the clockwise direction. With evolution continues, the vortex pattern appears with vortex-antivortex (V-AV) pairs. We divide the evolution process into three different stages. The first stage is the small V-AV nucleation stage. The second stage is the coarsing stage, which the small vortex will coarsing into bigger vortex and stabilized with no vortex domain changing at the end if there are no strain applied. The third stage is V-AV movement stage which starts from 10^4 evolution steps. Due to the elastic coupling effect, the vortex domain gradually changes to stripe domain.

Table 1
The parameters used in phase-field simulation [15].

a (eVÅ^{-2})	b (eVÅ^{-4})	c (eVÅ^{-6})	c'(eVÅ^{-6})	a_p(eVÅ^{-2})	g (eVÅ^{-4})	g'(eVÅ^{-4})
−2.626	3.375	0.117	0.108	0.866	1.945	9.931
s_Q^x(eV)	s_Q^y(eV)	s_Q^z(eV)	s_P^x (eV)	s_P^y (eV)	s_P^z(eV)	L
5.14	5.14	15.4	−8.88	−8.88	52.7	1