Numerical Modeling of Polarization Vortex Evolution in Strained Ferroelectrics

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Abstract

An evolution of ferroelectric vortex-antivortex lattices (FVL) in strained ferroelectric materials has been observed experimentally. The discovery of polar vortex textures revealed a new facet of nanoscale ferroelectric materials with prospects for new interaction pathways with chiral, topological, and photonic materials. As experiments and calculations show, FVL have a frequency response in the sub-THz range and thus, electromagnetic (EM) waves at these frequencies may interact with FVL. Therefore, extensive numerical studies of these structures are needed to exploit e.g. anomalous scattering responses and enhanced THz Kerr-type nonlinearity. In this contribution, we present a numerical model of vortex evolution in such material systems which is prepared to analyze (tailored) EM wave-matter interactions in the future.

1 Introduction

Ferroelectric materials are the electrical analogue to ferromagnetics, i.e., the polarization density (or electric flux density) as function of the electric field is forming a hysteresis. This is accompanied by a non-vanishing polarization density in the ground state when no external electric field is applied, similar to pyroelectrics. This behavior of ferroelectric (and pyroelectrics) materials follows from thermodynamic principles. For a body at constant temperature, this is the endeavor to minimize a systems free energy F [1].

The behavior of bulk ferroelectric materials is well known and exploited for decades in electronic and optical devices such as capacitors, ferroelectric memory, and optical modulators. However, recent experimental observations have demonstrated the existence of polarization vortex-antivortex lattices in strained layers of two different ferroelectric materials [2] with a periodicity p in the order of 10 nm, as schematically shown in **Fig. 1**. Experiments [3] and calculations [4] report a frequency response of these ferroelectric vortex-antivortex lattices (FVL) in the sub-THz regime (where the wavelength $\lambda \gg p$), caused by the eigenfrequency of formed discrete dipoles, which start oscillating when perturbed from their ground state due to an impinging electromagnetic (EM) wave [4], [5]. This observation indicates such materials to be useful in developing electronic devices operating in the sub-THz regime. In this context, analytical and numerical models of wave-matter interactions are required. As a first step towards this goal, we demonstrate the evolution of FVL in the time domain by minimizing the free energy.

2 Modeling

The dipole dynamics inside a FVL is governed by a second-order time-dependent Landau-Ginzburg-Devonshire (LGD) equation [6]

$$\alpha_{\mathrm{k}} \frac{\partial^2 P_\ell}{\partial t^2} + \frac{1}{L} \frac{\partial P_\ell}{\partial t} = -\frac{\delta F}{\delta P_\ell}, \tag{1}$$

where $\boldsymbol{P}$ is the polarization density, α_{k} and L are kinetic coefficients, t stands for the time, and index ℓ referes to the vector component in ℓ-direction. Based on the LGD theory, assuming centrosymmetry and using Einstein summation convention, the free energy functional F can be written as [3]

$$\begin{aligned} F = \int_V \Bigg\{ & \alpha_{ij} P_i P_j + \alpha_{ijk\ell} P_i P_j P_k P_\ell \\ & + \alpha_{ijk\ell mn} P_i P_j P_k P_\ell P_m P_n \\ & + \frac{1}{2} g_{ijk\ell} \frac{\partial P_i}{\partial x_j} \frac{\partial P_k}{x_\ell} + \frac{1}{2} c_{ijk\ell} \epsilon_{ij} \epsilon_{k\ell} \\ & - q_{ijk\ell} \epsilon_{ij} P_k P_\ell - \frac{1}{2} \varepsilon_b E_i E_i - E_i P_i \Bigg\} \mathrm{d}V, \end{aligned} \tag{2}$$

where α, g, c, q, ε_b are Landau coefficients, polarization gradient energy coefficients, elastic stiffness coefficients, electrosctrictive coupling coefficients, and the isotropic background permittivity, respectively, which are all material parameters. Here, ϵ_{ij} and E_i are elements of the mechanical strain tensor and the electric field vector, respectively, and the integration is performed over the entire volume of interest V.

Including mechanical effects of (2) in modified Landau coefficients [7] (indicated by a tilde), assuming the structure under investigation to be cubic-symmetric and invariant along the z-direction, as well as using Voigt-notation for the Landau coefficients, the free energy can be written compactly as [7]

$$\begin{aligned} F = \int \Bigg\{ & \tilde{\alpha}_1 \left(P_x^2 + P_y^2\right) + \tilde{\alpha}_{11} \left(P_x^4 + P_y^2\right) + \tilde{\alpha}_{12} P_x^2 P_y^2 \\ & + \tilde{\alpha}_{111} \left(P_x^6 + P_y^6\right) + \tilde{\alpha}_{112} \left(P_x^4 P_y^2 + P_x^2 P_y^4\right) \\ & + \frac{g_0}{2} \left[\left(\frac{\partial P_x}{\partial x}\right)^2 + \left(\frac{\partial P_x}{\partial y}\right)^2 + \left(\frac{\partial P_y}{\partial x}\right)^2 + \left(\frac{\partial P_y}{\partial y}\right)^2 \right] \\ & - \frac{1}{2} \varepsilon_b \left(E_x^2 + E_y^2\right) - E_x P_x - E_y P_y \Bigg\} \mathrm{d}V. \end{aligned} \tag{3}$$

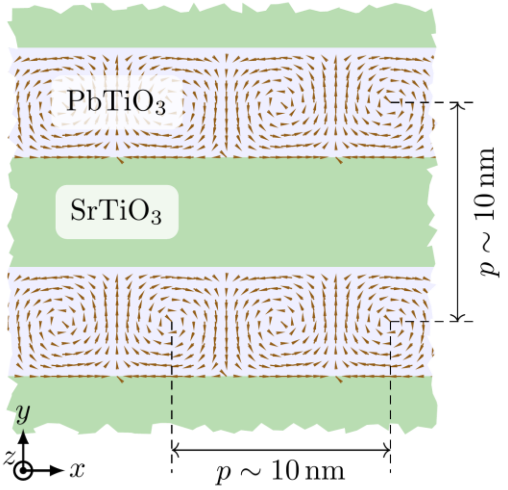

Figure 1: Schematical representation of a FVL composed of alternating layers of $PbTiO_3$ and $SrTiO_3$. The periodicity p of the polarization vortices evolving predominantly in the $PbTiO_3$ layer (indicated by the vector field) is in the order of 10 nm.

Typically (1) is solved assuming electro-quasistatic behavior in (2) within the domain of interest. However, in preparation to analyze the wave-matter interaction of an EM wave with the FVL, here we couple (1) to the governing equations of an EM wave, i.e. Maxwell equations

$$\nabla \times \boldsymbol{E} = -\mu_0 \frac{\partial \boldsymbol{H}}{\partial t}, \qquad (4a)$$

$$\nabla \times \boldsymbol{H} = \varepsilon_0 \frac{\partial \boldsymbol{E}}{\partial t} + \frac{\partial \boldsymbol{P}}{\partial t}, \qquad (4b)$$

where $\boldsymbol{H}$ is the magnetic field and ε_0 and μ_0 are the vacuum permittivity and permeability, respectively.

To model the vortex evolution in a system of alternating $PbTiO_3/SrTiO_3$ layers (cf. Fig. 1), we solve the coupled system of differential equations (1), (4a), and (4b) numerically using the finite-difference time-domain method (FDTD). At all boundaries, we applied periodic boundary conditions (BCs) (see Fig. 1) and to mimic the strain between the layers and to initiate the formation process, a fixed polarization density is forced at the interface between the two layers according to the second-order analytical solution in [7]. The material parameters are taken from [3] but we adapted the kinetic coefficients to accelerate the convergence. The vortex evolution process in a $PbTiO_3$layer is depicted in **Fig. 2** at the beginning of the process and in the steady state.

3 Conclusion and Outlook

In this paper, a method is presented to model the polarization vortex evolution of an FVL. By coupling the time-dependent LGD equation with Maxwell equations, the model is prepared to analyze the interaction of a FVL with an EM wave which we are currently investigating regarding to anomalous scattering responses and enhanced THz Kerr-type nonlinearity.

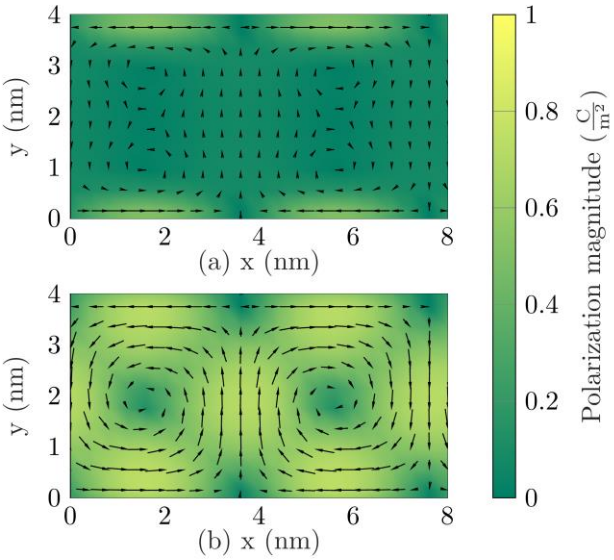

Figure 2: Evolution of polarization vortices in a $PbTiO_3$ layer. (a) depicts the polarization at the very beginning of the process and (b) the steady state.

Acknowledgements

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