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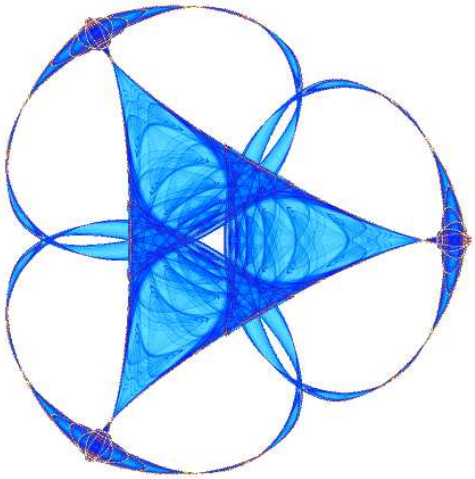

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Existence of Finitely Many Solution Branches and Nested Hysteresis Loops in Ferroelectric Materials

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Abstract

We consider Ginzburg-Landau energy for ferroelectric materials so as to study their structures during the polarization reversal process by an externally applied electric field. We accomplish very useful mathematical results which connect this problem with other situations in material sciences. Taking into account the Neumann boundary condition, we obtain inhomogeneous solutions and study hysteresis effects in the relation between polarization and applied field, which characterizes the ferroelectric phenomenon.

1 Introduction

In this article, we consider static configurations of Ginzburg-Landau energies of the polarization which plays an important role in the structure of ferroelectric materials. We study existence of finitely many nontrivial branches of equilibrium configurations of the energies by means of bifurcation analysis and investigate hysteresis loops which explain a relation between the polarization and an applied field. We reveal a way of understanding the polarization reversal process by way of nucleations.

In the study of soft matter systems, there are many materials exhibiting *ferroelectricity* which refers to the permanent or spontaneous polarization. Due to the appearance of the polarization, the structures of these materials are so complicated that a great deal of effort has been invested from the viewpoints of both mathematics and physics. A various different patterns of the polarization have been observed in the physics literature [10, 13, 17]. There are two typical types among

others, which are called *ferroelectric* and *antiferroelectric phases*. In the ferroelectric phase, the polarization prefers to be aligned to one direction so that the net polarization is not zero. But if the material in the antiferroelectric phase, the net polarization is zero because two opposite neighboring polarizations are canceled. If an electric field is applied in such phases, the dipoles which are already pointed in the direction of the applied field will remain so aligned, but those which are oriented in the opposite direction to the field will tend to reverse their orientation. This results in nucleations and domain walls, and a relation between polarization and applied field, i.e., *a hysteresis loop*, which is the most important characteristic of ferroelectric materials. For a small electric field applied in the direction of increasing the net polarization, there is almost linear relationship between the polarization and electric field because the field is not large enough to switch all of the domain. When we increase the strength of the electric field, the net polarization will increase rapidly until the state in which the polarization is aligned to the direction of the field. In fact, at a certain critical field strength a discontinuous change in polarization occurs. This critical field is necessary to induce the uniform state. We now decrease the strength of the electric field and then apply it in the opposite direction in order to get a hysteresis loop. The problem then arises of *how to specify the critical fields that can induce the switching and how to understand the effect of the polarization reversal process on the hysteresis loop during the switching by the applied field.*

In order to understand the process of the polarization reversal, we treat the net polarization as a bifurcation parameter, which is the key idea in the analysis, and investigate nontrivial branches of equilibrium configurations with a constraint on the net polarization. We use these branches of equilibrium configurations in order to prove existence of finitely many nested hysteresis loops in the sense that one loop lying inside a loop contains another loop, exhibiting very interesting phenomena which have not been reported in the literature. We understand that these nested hysteresis loops explain complex periodic structures of ferroelectric materials. It is also pointed out that critical fields inducing nucleations depend on the net polarization and can be calculated in terms of the polarization and the net polarization. As a consequence, we also obtain hysteresis loop between strain and applied field.

In our analysis, the coefficient of the polarization gradient plays a key role in occurring nucleations, which correspond to nontrivial solutions bifurcating at critical values of the total polarization. The smaller the value of the gradient coefficient of polarization, the larger the number of bifurcation points. There are lots of works regarding hysteresis loops for dynamical models in many different contexts. Among many others, dynamic models of a viscoelastic bar were studied by A.

Vainchtein, P. Rosakis, and T. Healey [21, 22, 23]. But the present work concerns static problems and the main interest of this paper is to study behaviors of solutions between two ferroelectric (uniform) states.

Throughout this paper, we denote by u the polarization vector and consider a system of a ferroelectric material occupying a domain Ω, which is governed by the Ginzburg-Landau energy

$$\int_\Omega \left\{ \frac{\varepsilon^2}{2}|\nabla u|^2 + f(u) \right\} dx, \tag{1}$$

where $\varepsilon > 0$ is a fixed number and $f(u)$ is given a function. In our analysis, we consider two types of energies for $f(u)$

$$\begin{cases} \text{Type I:} & f(u) = \frac{1}{6}u^2[(u^2-1)^2 - \alpha] & \text{for antiferroelectrics,} \\ \text{Type II:} & f(u) = \frac{1}{4}(u^2-1)^2 & \text{for ferroelectrics,} \end{cases}$$

where α is a constant depending on the temperature. Many works related with the Ginzburg-Landau energy with type II have been done by many authors, so that it is impossible to list all of them. Among them, J. Carr, M. Gurtin, and M. Slemrod [4] studied global minimizers of the energy functional, and $\Gamma-$ convergence analysis of the energy minimizers are found in [15, 16, 20]. Not many results related with Type I are available in the literature.

An example of the problem (1) arises from the study of ferroelectric solids. In particular, many important ferroelectric materials exhibit *perovskite* structures. In the perovskite family, the prototype phase is cubic, and all other phases including tetragonal [10] can be derived from the cubic phase by way of small lattice distortions or removal of some atoms. Among many others, W. Cao and L. E. Cross [3] have developed the Landau-Devonshire continuum model based on the symmetry requirement (see section 2).

The paper is organized as follows. We introduce three dimensional Landau-Devonshire model of materials in the perovskite family and obtain a one dimensional model as in (1) with a special structure in section 2. In section 3, we prove existence of finitely many nontrivial branches of equilibrium configurations of the problem by means of local and global bifurcation theories. We then obtain finitely many hysteresis loops, and present examples in section 4. We discuss conclusion in section 6.

2 Landau-Devonshire Model of Ferroelectric Solids

In this section, we introduce the Landau-Devonshire model developed by [3] in order to study phase transitions in ferroelectric solids. Using

the idea of Landau for a second order phase transition, the free energy has terms of a polynomial expansion of the polarization, which is the primary order parameter in a ferroelectric phase transition. Most of all known ferroelectric materials have a property that the spontaneous strain occurs due to the formation of the spontaneous polarization. In order to account for this property, the total energy also includes linear elastic energy for mechanical displacement and the coupling term with the polarization in the Landau-Devonshire model.

2.1 Energy functional of the continuum model

We denote the material measure of the polarization by $\mathbf{P}$ and $\mathbf{v}$ denotes the displacement of the material particle. We also define the symmetry part of the gradient of the displacements by

$$e(\mathbf{v}) = \frac{1}{2}\left(\nabla\mathbf{v} + \nabla\mathbf{v}^T\right).$$

The free energy of the Landau-Devonshire model for a ferroelectric crystal [3] is given by

$$F(\mathbf{P}, \nabla\mathbf{P}, \nabla\mathbf{v}) = F_{el}(\nabla\mathbf{v}) + F_c(\mathbf{P}, \nabla\mathbf{v}) + F_P(\mathbf{P}, \nabla\mathbf{P}),$$

where

$$\begin{aligned}
F_{el}(\nabla\mathbf{v}) &= \frac{C_{11}}{2}\sum_{i=1}^{3} e_{ii}(\mathbf{v})^2 + C_{12}\sum_{i<j} e_{ii}(\mathbf{v})e_{jj}(\mathbf{v}) + C_{44}\sum_{i\neq j} e_{ij}(\mathbf{v})^2,\\
F_c(\mathbf{P}, \nabla\mathbf{v}) &= -q_{11}\sum_{i=1}^{3} e_{ii}(\mathbf{v})P_i^2 - q_{12}\sum_{i=1}^{3}\left(\sum_{j\neq i} e_{ii}(\mathbf{v})P_j^2\right) - 2q_{44}\sum_{i<j} e_{ij}(\mathbf{v})P_iP_j,\\
F_P(\mathbf{P}, \nabla\mathbf{P}) &= \frac{1}{2}g_{11}\sum_{i=1}^{3} P_{i,i}^2 + g_{12}\sum_{i<j} P_{i,i}P_{j,j} + \frac{1}{2}g_{44}\sum_{i\neq j}(P_{i,j}+P_{j,i})^2\\
&\quad + a_1\sum_{i=1}^{3} P_i^2 + a_{11}\left(\sum_{i=1}^{3} P_i^2\right)^2 + a_{12}\sum_{i<j} P_i^2P_j^2\\
&\quad + a_{111}\sum_{i=1}^{3} P_i^6 + a_{112}\sum_{i=1}^{3} P_i^4\left(\sum_{i\neq j} P_j^2\right) + a_{123}P_1^2P_2^2P_3^2,
\end{aligned}$$

where $\mathbf{P} = (P_1, P_2, P_3), P_{i,j} = \frac{\partial P_i}{\partial x_j}$.

We assume that

$$\begin{cases} a_{11} > 0, \quad a_{111} > 0, \quad C_{11} > 0, \quad C_{44} > 0,\\ g_{11} > 0, \quad g_{44} > 0, \quad g_{12} < 0, \end{cases}$$

and $a_1 = a_0(T - T_0)(a_0 > 0)$ will change sign with temperature. The physical meaning of the coefficients can be found in [3] and all coefficients except a_1 are assumed to be independent of temperature

The energy F_{el} is the linear elastic energy associated with the mechanical deformation, F_P is the bulk free energy for the polarization together with the gradient contribution, and finally F_c is the coupling between the elastic and the polarization effects in the free energy.

The corresponding Euler-Lagrange equations are given by

$$\begin{cases} \frac{\partial}{\partial x_j}\left(\frac{\partial F}{\partial P_{i,j}}\right) - \frac{\partial F}{\partial P_i} = 0, \\ \sigma_{ij,j} = \frac{\partial}{\partial x_j}\left(\frac{\partial F}{\partial e_{ij}}\right) = 0. \end{cases} \tag{2}$$

It is easy to see that the equations (2) allow for two types of homogeneous solutions:

$$\begin{cases} \mathbf{P} = (0,0,0); \\ \mathbf{P} = (\pm P^*, 0, 0), (0, \pm P^*, 0), (0, 0, \pm P^*), \end{cases}$$

for some fixed number P^*. The first solution represents the cubic phase, in which the polarization is zero, and the latter three solutions describe the tetragonal phases where the polarization will point along a cube edge. The corresponding homogeneous strain can be also calculated easily from (2) (see [3]).

2.2 A one dimensional model of ferroelectric solids

As in the study of 180° domains [3], we assume that $\mathbf{P} = (0, 0, P)$. Then the energy associated with the polarization is given by

$$\begin{aligned} F_P = \frac{1}{2} g_{11} \left(\frac{\partial P}{\partial z}\right)^2 + \frac{g_{44}}{2} \left[\left(\frac{\partial P}{\partial x}\right)^2 + \left(\frac{\partial P}{\partial y}\right)^2\right] \\ + a_1 P^2 + a_{11} P^4 + a_{111} P^6. \end{aligned} \tag{3}$$

In fact, the energy in (3) is a typical free energy for a crystal in the $D_{6d} - C_{6v}$ or $D_{3d} - C_{3v}$ phase transitions [2]. Assuming that P and $\mathbf{v}$ depend only on x, the equations in (2) can be written as

$$\begin{cases} g_{44} \frac{d^2 P}{dx^2} = 2a_1 P + 4a_{11} P^3 + 6a_{111} P^5 - 2q_{12} P \frac{dv_1}{dx}, \\ C_{11} \frac{dv_1}{dx} - q_{12} P^2 = s, \end{cases} \tag{4}$$

for any constant s. Plugging the second equation into the first one, we obtain

$$g_{44} \frac{d^2 P}{dx^2} = aP + bP^3 + cP^6, \tag{5}$$

where a, b, c are

$$a = 2\left(a_1 - \frac{q_{12}s}{C_{11}}\right), b = 2\left(2a_{11} - \frac{q_{12}^2}{C_{11}}\right), c = 6a_{111}.$$

We notice that the constant s will be determined by boundary conditions for P and $\mathbf{v}$. The equation in (5) is the Euler-Lagrange equation for the energy functional

$$\frac{1}{2}\int_0^1 \left\{ g_{44}\left(\frac{dP}{dx}\right)^2 + aP^2 + bP^4 + cP^6 \right\} dx, \tag{6}$$

with the Neumann boundary condition for P.

Assuming $b < 0$, let

$$P_0^2 = \frac{|b|}{2c}, \alpha = \frac{b^2 - 4ac}{b^2}, \varepsilon^2 = \frac{g_{44}}{2cP_0^4}.$$

Divide the energy in (6) by $3cP_0^6$ to get

$$\int_0^1 \left\{ \frac{\varepsilon^2}{2}(u'(x))^2 + \frac{1}{6}u^2[(u^2-1)^2 - \alpha] \right\} dx,$$

which we study in later sections.

3 Finitely Many Branches of Equilibrium Configurations

Throughout this paper, we assume that a sufficiently small number $\varepsilon > 0$ is fixed.

We study equilibrium configurations of the problem

$$\begin{cases} \mathbb{E}(u) = \int_0^1 \left\{ \frac{\varepsilon^2}{2}(u'(x))^2 + f(u) \right\} dx \\ \text{subject to} \\ u'(0) = u'(1) = 0, \ \int_0^1 u\, dx = \lambda, \quad \lambda \in \mathbf{R}, \end{cases} \tag{7}$$

where $f(u)$ is given a function.

Definition 3.1. *The problem (7) is called*

$$\begin{cases} \textit{antiferroelectrics with } \alpha & \textit{if } f(u) = \frac{1}{6}u^2[(u^2-1)^2 - \alpha], \\ \textit{ferroelectrics} & \textit{if } f(u) = \frac{1}{4}(u^2-1)^2. \end{cases}$$

The corresponding Euler-Lagrange equation is

$$\begin{cases} -\varepsilon^2 u''(x) + f'(u) = \int_0^1 f'(u(s))\, ds \text{ in } [0,1], \\ u'(0) = u'(1) = 0, \ \int_0^1 u\, dx = \lambda. \end{cases} \tag{8}$$

First, we observe that $u = \lambda$ is a solution for any $\lambda \in \mathbf{R}$. Introducing a new variable w by $u = w + \lambda$, the equation (8) is equivalent to

$$\begin{cases} -\varepsilon^2 w''(x) + f'(w(x) + \lambda) = \int_0^1 f'(w(s) + \lambda)\, ds \text{ in } [0,1], \\ w'(0) = w'(1) = 0, \ \ \int_0^1 w\, dx = 0. \end{cases} \tag{9}$$

Let

$$\mathcal{X} = \left\{ w \in C^2[0,1] : w'(0) = w'(1) = 0, \int_0^1 w\, dx = 0 \right\},$$
$$\mathcal{Z} = \left\{ z \in C^0[0,1] : \int_0^1 w\, dx = 0 \right\},$$

where $C^n[0,1]$ is the set of all functions from $[0,1]$ from $\mathbf{R}$ whose jth derivatives ($0 \le j \le n$) are continuous. Clearly, $\mathcal{X}$ and $\mathcal{Y}$ are not empty and Hilbert spaces with the inner product $<,>_2$, defined by

$$< u, v >_2 = \int_0^1 u(s)v(s)\, ds$$

for two functions u and v.

Define $G : \mathcal{X} \times \mathbf{R} \to \mathcal{Z}$ by

$$G(w, \lambda) = -\varepsilon^2 w'' + f'(w + \lambda) - \int_0^1 f'(w(s) + \lambda)\, ds. \tag{10}$$

Then $G(0, \lambda) = 0$ for any $\lambda \in \mathbf{R}$.

The linearized equation of (9) at $(0, \lambda)$ is given by

$$\begin{cases} -\varepsilon^2 w'' + f''(\lambda) w = 0 \text{ in } [0,1], \\ w'(0) = w'(1) = 0, \ \ \int_0^1 w\, dx = 0 \end{cases} \tag{11}$$

The solution space of the linearized equation is spanned by $\cos k\pi x$ if there exists $k \in \mathbb{N}$ satisfying

$$\varepsilon^2 k^2 \pi^2 = -f''(\lambda). \tag{12}$$

Since ε is sufficiently small, there always exists a pair of λ and k satisfying (12).

The number of real roots of (12) depends on $f(u)$ and α. We discuss them as follows.

Antiferroelectrics with α. Let $f(u) = \frac{1}{6}u^2\left[(u^2-1)^2 - \alpha\right]$.

1. $\alpha \le -\frac{7}{5}$: There is no bifurcation point.
2. $-\frac{7}{5} < \alpha < 1$: Let K be the largest integer satisfying

$$\varepsilon^2 k^2 \pi^2 < \frac{7}{15} + \frac{\alpha}{3}, \text{ for } 1 \le k \le K. \tag{13}$$

Then for each $1 \le k \le K$, there are four real roots of (12), two pairs of which have opposite signs. We denote them by

$$\pm\lambda_k^i, (i = 1, 2), \;\; 0 < \lambda_k^1 < \sqrt{\frac{2}{5}} < \lambda_k^2. \tag{14}$$

3. $\alpha > 1$: Let K_0 be the largest integer satisfying

$$\varepsilon^2 k^2 \pi^2 < \frac{1}{3}(\alpha - 1) \text{ for } 1 \le k \le K_0. \tag{15}$$

   (a) If $1 \le k \le K_0$, then the equation (12) has two roots with opposite signs, which we label, $\pm\lambda_k$, $\lambda_k > \sqrt{\frac{2}{5}}$.

   (b) If $K_0 < k \le K$, then the equation (12) has four roots satisfying (14). We keep the same notation as in (14).

Ferroelectrics. Let $f(u) = \frac{1}{4}(u^2 - 1)^2$ and M be the largest integer satisfying

$$k^2 < \frac{1}{\varepsilon^2 \pi^2}. \tag{16}$$

Then for each $1 \le k \le M$, the equation (12) have two roots with opposite signs, and we label them by

$$\pm\eta_k, \;\; \eta_k = \sqrt{\frac{1 - \varepsilon^2 k^2 \pi^2}{3}}. \tag{17}$$

Convention. From now on, we keep all notations of roots discussed above for the rest of this paper.

3.1 Local bifurcation analysis

Now, let λ_0 denote one of λ satisfying (12) with corresponding to k, and L_{λ_0} be the linearized operator, $D_w G(0, \lambda_0)$, of G at $(0, \lambda_0)$. Then we obtain

$$\dim \ker L_{\lambda_0} = \; \text{codim } L_{\lambda_0} = 1.$$

In particular, $L_{\lambda_0} = -\varepsilon^2 w'' + f''(\lambda_0) w$ is a self-adjoint operator. By Lyapunov-Schmidt reduction, there exist spaces $\mathcal{X}_0$ and $\mathcal{Z}_0$ such that

$$\mathcal{X} = \ker L_{\lambda_0} \oplus \mathcal{X}_0, \qquad \mathcal{Z} = \mathcal{R}(L_{\lambda_0}) \oplus \mathcal{Z}_0.$$

where $P : \mathcal{X} \to \ker L_{\lambda_0}$ and $Q : \mathcal{Z} \to \mathcal{Z}_0$ are continuous projection maps. By direct computations, we establish

$$\ker L_{\lambda_0} = \text{span}\{\phi_k\}, \qquad \phi_k(x) = \sqrt{2} \cos k\pi x,$$
$$D^2_{w\lambda} F(0, \lambda_0)\phi_k \notin \mathcal{R}(L_{\lambda_0}).$$

It is also easy to see that

$$\mathcal{R}(L_{\lambda_0}) = \left\{ v \in \mathcal{Z} : \int_0^1 v(s)\phi_k(s)\,ds = 0 \right\},$$
$$\mathcal{Z}_0 = \text{ span } \{v_0^*\} = \mathcal{R}(L_{\lambda_0})^\perp = \ker L_{\lambda_0}^*,$$

where $v_0^* = \phi_k$. Furthermore, by Lyapunov Reduction, there exists a continuous map $\phi : \mathcal{U} \times (a,b) \to \mathcal{W}$ satisfying

$$QG(w + \phi(w,\lambda),\lambda) = 0 \text{ for all } (w,\lambda) \in \mathcal{U} \times (a,b), \;\; \phi(0,\lambda_0) = 0, \tag{18}$$

where $\mathcal{U}, (a,b)$ and $\mathcal{W}$ are open neighborhoods of $0, \lambda_0$, and w_0 in $\ker(L), \mathbf{R}$, and $\mathcal{X}_0$ respectively.

For all $(w,y) \in \mathcal{U} \times (a,b)$, we define Φ by

$$\Phi(v,\lambda) = QG(w + \phi(w,\lambda),\lambda). \tag{19}$$

By local bifurcation theorem [5], there exists a local nontrivial solution curve $\mathcal{S}$ in $\mathcal{X} \times \mathbf{R}$ emanating from $(0,\lambda_0)$ such that all solutions of $G(w,\lambda) = 0$ in a neighborhood of $(0,\lambda_0)$ are either on the trivial line or on the nontrivial curve $\mathcal{S}$. If $\mathcal{S}$ is parameterized by s near $(0,\lambda_0)$

$$\mathcal{S} : \{(w(s),\lambda(s)) : s \in (-\delta,\delta)\} \text{ for some } \delta > 0, \tag{20}$$

then $w(s)$ behaves like $s\phi_k + \omega(s)$ for some ω satisfying

$$\omega(0) = \dot{\omega}(0) = \frac{d\omega}{ds}\Big|_{s=0} = 0.$$

Lemma 3.2. *Let $\mathcal{S}$ be parameterized by s in (20). Then*

$$\dot{\lambda}(0) = 0, \qquad \ddot{\lambda}(0) = -\frac{1}{6}\left[\frac{3f^{(4)}(\lambda_0)}{f'''(\lambda_0)} + \frac{f'''(\lambda_0)}{f''(\lambda_0)}\right]. \tag{21}$$

Proof. By direct computations with using formulas in[9, pp. 31], we obtain

$$< D^2_{w\lambda}G(0,\lambda)v_0, v_0^* >= f'''(\lambda),$$
$$D^2_{ww}G(0,\lambda)[v_0,v_0] = f'''(\lambda)v_0^2 - f'''(\lambda)\int_0^1 v_0^2\,dx = f'''(\lambda)(v_0^2 - 1),$$
$$D^3_{www}G(0,\lambda)[v_0,v_0,v_0] = f^{(4)}(\lambda)v_0^3.$$

Furthermore, we have

$$< D_{ww}G(,\lambda)[v_0,v_0], v_0^* >= \int_0^1 \left\{f'''(\lambda)v_0^3 - f'''(\lambda)v_0\right\}\,dx = 0,$$
$$\dot{\lambda}(0) = -\frac{< D^2_{ww}G(0,\lambda)[v_0,v_0], v_0^* >}{< D_{w\lambda}G(0,\lambda)v_0, v_0^* >} = 0,$$

so that $D^2_{ww}G(0,\lambda)[v_0,v_0] \in \mathcal{R}(D_wG(0,\lambda)) = R(L)$. By Lyapunov-Schmidt reduction, we have

$$D^2_{ww}G(0,\lambda)[v_0,v_0] = (I-Q)D^2_{ww}G(0,\lambda)[v_0,v_0].$$

Next, we recall (see [12, pp. 19] or [9, pp. 33]) that

$$\ddot{\lambda}(0) = -\frac{1}{3}\frac{< D^3_{vvv}\Phi(0,\lambda)[v_0,v_0,v_0], v_0^* >}{< D^2_{x\lambda}G(0,\lambda)v_0, v_0^* >},$$

where Φ is defined in (19) and

$$\begin{aligned} &D^3_{vvv}\Phi(0,\lambda_0)[v_0,v_0,v_0] = QD^3_{vvv}G(0,\lambda_0)[v_0,v_0,v_0] \\ &-3QD^2_{xx}G(0,\lambda_0)\left[v_0,(I-P)L^{-1}(I-Q)D^2_{vv}G(0,\lambda)[v_0,v_0]\right]. \end{aligned}$$

In order to compute $\ddot{\lambda}(0)$, we need to find $L^{-1}D^2_{ww}G(0,\lambda)[v_0,v_0] = L^{-1}[f'''(\lambda)(v_0^2-1)]$ so that we are able to get

$$(I-P)L^{-1}D^2_{ww}G(0,\lambda)[v_0,v_0].$$

Let w be a solution in $\mathcal{X}$ satisfying

$$Lw = f'''(\lambda)(v_0^2-1), w'(0) = w'(1) = 0, \int_0^1 w\,dx = 0.$$

It is easy to get

$$w(x) = -\frac{f'''(\lambda)}{3f''(\lambda)}\cos 2k\pi x.$$

Since $w(x) = (I-P)w(x)$, we have

$$\begin{aligned} &(I-P)L^{-1}(I-Q)D^2_{ww}G(0,\lambda)[v_0,v_0] = \frac{f'''(\lambda)}{3\varepsilon^2k^2\pi^2}\cos 2k\pi x, \\ &QD^2_{ww}G(0,\lambda)[v_0,w] = \frac{[f'''(\lambda)]^2}{3\varepsilon^2k^2\pi^2}v_0\cos 2k\pi x. \end{aligned}$$

We then obtain

$$\begin{aligned} &D^3_{vvv}\Phi(0,\lambda_0)[v_0,v_0,v_0] = f^{(4)}(\lambda)v_0^3 - \frac{[f'''(\lambda)]^2}{\varepsilon^2k^2\pi^2}v_0\cos 2k\pi x, \\ &< D^3_{vvv}\Phi(0,\lambda_0)[v_0,v_0,v_0], v_0^* >= \frac{3f^{(4)}(\lambda)}{2} - \frac{[f'''(\lambda)]^2}{2\varepsilon^2k^2\pi^2}. \end{aligned}$$

Since $\varepsilon^2k^2\pi^2 = -f''(\lambda)$, we get

$$\ddot{\lambda}(0) = -\frac{1}{6}\left[\frac{3f^{(4)}(\lambda)}{f'''(\lambda)} + \frac{f'''(\lambda)}{f''(\lambda)}\right].$$

□

Using the formula in lemma 3.2, we evaluate $\ddot{\lambda}(0)$ at each point to decide type of bifurcation points. If $(0,\lambda)$ satisfies $\ddot{\lambda}(0) > 0$($\ddot{\lambda}(0) < 0$), then it is called *supercritical* (*subcritical*).

Corollary 3.3. *All bifurcation points are pitchforks. In particular, if* $(0,\lambda_0)$ *is a bifurcation point, then it is*

$$\begin{cases} \textit{subcritical} & \textit{if } \lambda_0 \in \{\lambda_k, -\lambda_k^1, \lambda_k^2, -\eta_k\}, \\ \textit{supercritical} & \textit{if } \lambda_0 \in \{-\lambda_k, \lambda_k^1, -\lambda_k^2, \eta_k\}. \end{cases} \tag{22}$$

It is not hard to check the following properties of $D_w G(0,\lambda)$ which are useful to understand local stabilities of nontrivial solution branches. *Antiferroelectrics with* α:

1. $-\frac{7}{5} < \alpha < 1$: The kth eigenvalue of $D_w G(0,\lambda)$ is

$$\begin{cases} \text{negative} & \text{if } \lambda \in (-\infty, -\lambda_k^2) \cup (-\lambda_k^1, \lambda_k^1) \cup (\lambda_k^2, \infty), \\ \text{positive} & \text{if } \lambda \in (-\lambda_k^2, -\lambda_k^1) \cup (\lambda_k^1, \lambda_k^2), \end{cases}$$

   where $1 \le k \le K$.

2. $\alpha > 1$:
   (a) If $1 \le k \le K_0$, then kth eigenvalue of $D_w G(0,\lambda)$ is

$$\begin{cases} \text{negative} & \text{if } \lambda \in (-\infty, -\lambda_k) \cup (\lambda_k, \infty), \\ \text{positive} & \text{if } \lambda \in (-\lambda_k, \lambda_k). \end{cases}$$

   (b) If $K_0 < k \le K$, then the kth eigenvalue of $D_w G(0,\lambda)$ is

$$\begin{cases} \text{negative} & \text{if } \lambda \in (-\infty, -\lambda_k^2) \cup (-\lambda_k^1, \lambda_k^1) \cup (\lambda_k^2, \infty), \\ \text{positive} & \text{if } \lambda \in (-\lambda_k^2, -\lambda_k^1) \cup (\lambda_k^1, \lambda_k^2). \end{cases}$$

Ferroelectrics: For $1 \le k \le K$, the kth eigenvalue of $D_w G(0,\lambda)$ is

$$\begin{cases} \text{negative} & \text{if } \lambda \in (-\infty, -\eta_k) \cup (\eta_k, \infty), \\ \text{positive} & \text{if } \lambda \in (-\eta_k, \eta_k). \end{cases}$$

3.2 Global bifurcation analysis and existence of finitely many branches of equilibrium configurations

In this section, we study the global behavior of nontrivial solution branches emanating from bifurcation points. Specifically, we prove existence of finitely many closed curves consisting of these nontrivial branches. Moreover, the number of these closed solutions curves goes to ∞ as $\varepsilon \to 0$.

Theorem 3.4. *Let* $\mathcal{M}_k$ *be a maximal connected subset of the closure of the nontrivial solution branch bifurcating from* $(0,\lambda_0)$ *where* $\lambda_0 \in \{\pm\lambda_k, \pm\lambda_k^i (i = 1,2), \pm\eta_k\}$. *Then* $\mathcal{M}_k$ *is bounded and* $\mathcal{M}_l \cap \mathcal{M}_l = \emptyset$ *if* $k \ne l$.

Proof. By properties of the function f'(either antiferroelectrics with α or ferroelectrics), there exists $M_1 > 0$ such that

$$\begin{cases} f' \text{ is increasing on } [M_1, \infty), \\ f' \text{ is decreasing on } (-\infty, -M_1] \ , \\ f'(-M_1) < f'(s) < f'(M_1) \text{ for all } -M_1 < s < M_1. \end{cases} \tag{23}$$

Let (w, λ) be any solution pair in $\mathcal{X} \times \mathbf{R}$ which lies on $\mathcal{S}$. Since $w \in C^2[0,1]$, w has the absolute minimum at x_0 and absolute maximum at x_1 for some $x_0, x_1 \in [0,1]$. Since $\int_0^1 w(s)\, ds = 0$, we get $w(x_0) < 0 < w(x_1)$ and w satisfies the Euler-Lagrange equation

$$-\varepsilon^2 w''(x) + f'(w(x) + \lambda) = \int_0^1 f'(w(s) + \lambda)\, ds \text{ for all } x \in [0,1].$$

This implies the following inequality

$$f'(w(x_1) + \lambda) \le f'(w(x_0) + \lambda).$$

From (23), we get $-M_1 \le w(x_0) + \lambda \le \lambda$ and $\lambda \le w(x_1) + \lambda \le M_1$ so that $|\lambda| \le M_1$ and $||w||_\infty \le M_1$. From the Euler-Lagrange equation, $||w'||_\infty$ and $||w''||_\infty$ are bounded by $\frac{C_1}{\varepsilon^2}$ where C_1 depending on M. Hence, $|\lambda| + ||w||_{C^2[0,1]} \le D$ where D depends only on M, ε and therefore $\mathcal{M}_k$ is bounded. Now, any element w in $\mathcal{S}$ has exactly k zeros, each of which is simple (characterized by $\cos k\pi x$). Let $\mathcal{Z}_j$ is the set of all functions in $\mathcal{X}$ which have exactly j zeros which are simple. By the standard argument [1, pp.164], the sets $\mathcal{Z}_j$ are disjoint. This completes the proof. □

Let $(0, \lambda)$ be a bifurcation point corresponding to k. We rewrite the Euler-Lagrange equation (9) by

$$w - \frac{1}{\varepsilon^2} \left(\frac{d^2}{dx^2} \right)^{-1} \left[f'(w + \lambda) - \int_0^1 f'(w(s) + \lambda) \right] = 0. \tag{24}$$

Choose $\mathcal{Y} = \mathcal{Z}$ and define $F : \mathcal{Y} \times \mathbf{R} \to \mathcal{Y}$ by

$$F(w, \lambda) = w - \frac{1}{\varepsilon^2} \left(\frac{d^2}{dx^2} \right)^{-1} \left[f'(w + \lambda) - \int_0^1 f'(w(s) + \lambda) \right].$$

Then $\frac{1}{\varepsilon^2}$-term is a compact operator, and $D_w F(0, \lambda)$ has an odd crossing number at λ because its kernel has the dimension 1. So we can apply the Global Bifurcation Theorem [6, 8].

Now, we observe how the number of real roots of (12) changes by the value of α in the antiferroeletrics with α. From (13)-(15), the critical case occurs when $\alpha = 1$. For a fixed ε, the number of real roots of (12) corresponding to $k = K_0$ jumps from 2 to 4 by decreasing the

value of α because of (15). The new roots are very close to 0 near the transition. This implies that two roots $\pm\lambda_k^1$ appear or disappear at the same time according the value of α.

For $1 \le k \le K_0$, let $\mathcal{M}_k$ be the maximal connected subset of the closure of the nontrivial solution curve bifurcating from $(0, \lambda_k)$. By the Global bifurcation theorem, the curve $\mathcal{M}_k$ connects with $(0, -\lambda_k)$ because $\mathcal{M}_k$ cannot contain $\pm\lambda_j (j \neq k)$. Remember that the absolute value of these roots is always great than $\sqrt{\frac{2}{5}}$. By the principle of exchange of stability [7], two bifurcation points $(0, \pm\lambda_j^1)(K_0 < j \le K)$ cannot be connected. This implies that a single closed curve composed of two nontrivial solution branches bifurcating from $(0, \pm\lambda_{K_0})$ is split into two branches when we decrease the value of α. As a summary, we conclude the following theorem.

Theorem 3.5. *Let a sufficiently small ε be given.*

1. *(Antiferroelectric with α)*
   (a) *If $1 \le k \le K_0$, then there is a single closed nontrivial solution curve (labeled $\mathcal{C}^s_{k,\mathcal{A}}$), which connects $(0, \lambda_k)$ with $(0, -\lambda_k)$.*
   (b) *If $K_0 < k \le K$, then there are two closed nontrivial solution curves (labeled $\mathcal{C}^{d,\pm}_{k,\mathcal{A}}$) , which connect $(0, \pm\lambda_k^1)$ with $(0, \pm\lambda_k^2)$ respectively.*
2. *(Ferroelectrics) For each $1 \le k \le M$, there exists a single closed solution curve, $\mathcal{C}^s_{k,\mathcal{F}}$, comprised of two nontrivial solution branches bifurcating from $(0, \pm\eta_k)$.*

As a consequence, we obtain the corollary.

Corollary 3.6. *For a sufficiently small $\varepsilon > 0$, there are finitely many local minimizers of the problem (7) for each λ, where*

$$\begin{cases} \lambda \in (-l_-, l_-) & \text{for antiferroelectrics with } \alpha, \\ \lambda \in (-l_+, l_+) & \text{for ferroelectrics }, \end{cases}$$

with

$$l_-^2 = \frac{1}{5}\left[2 + \sqrt{4 - 5\varepsilon^2\pi^2 - \frac{5}{3}(1-\alpha)}\right], \quad l_+^2 = \frac{1}{3}(1 - \varepsilon^2\pi^2), \quad l_\pm > 0.$$

Moreover, the number of local minimizers goes to ∞ as $\varepsilon \to 0$ for $\lambda = \lambda^*$, where

$$\lambda^* = \begin{cases} \pm\sqrt{\frac{2}{5}} & \text{for antiferroelectrics with } \alpha, \\ 0 & \text{for ferroelectrics.} \end{cases}$$

Schematic shapes of the nontrivial solution branches obtained in this section are described in Figures 1 - 2. It should be addressed that figures drawn here are not real solution curves. Real solution curves may be more complicated than the ones in Figures 1 - 2. As we see from Figures 1(R)-2(L), there exists a pair of two closed curves inside a single closed curve when ε is very small and $\alpha > 1$. Outer curves are pretty much similar to the curves in ferroelectrics as in Figure 2(R). But there always exists a pair of curves inside these curves. This explains that the structures of antiferroelectric materials are more complex than the ones in ferroelectrics. So, antiferroelectric materials show a rich variety of potential applications in practice.

In Figure 1(L) and 1(M), we take $K = 3$ and $-\frac{7}{5} < \alpha < 1$. The Figure 1(L) is a schematic curve for α being away from 1, while Figure 1(M) is drawn when α is close to 1. We take $k_0 = 1, K = 3$ and $\alpha(> 1)$ close to 1 in Figure 1(R). In Figure 2(L), we take $K = 4, k_0 = 1$, and $\alpha(> 1)$ (away from 1). The Figure 2(R) shows schematic solution branches for $K = 3$.

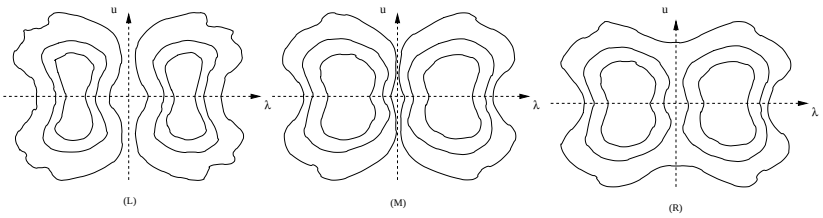

Figure 1: Antiferroelectrics

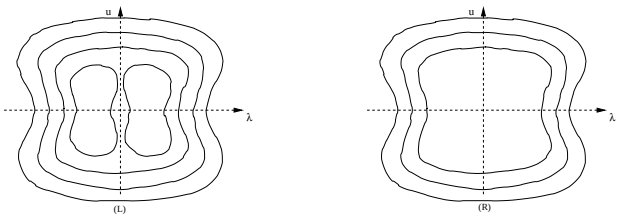

Figure 2: (L): Antiferroelectrics, (R): Ferroelectrics

After the present work was finished, professor R. V. Kohn informed the author that theorem 3.5 also remains true for two dimensional domains by an earlier work done by H. Kielhöfer [11].

4 Hysteresis Loops in Ferroelectric Materials

In this section, we use theorem 3.5 to prove existence of finitely many hysteresis loops of the polarization with respect to applied fields. We also present examples for applications with some experimental data.

4.1 Existence of finitely many nested hysteresis loops between the polarization and applied fields

In the present section, we prove our main theorem of existence of finitely many nested hysteresis loops.

Again, we consider a very simple case that the polarization depends only on x, and the following problem is given by

$$\begin{cases} \mathbb{E}(u) = \frac{1}{2}\int_0^1 \left\{\varepsilon^2 (u'(x))^2 + f(u) - Eu\right\}\, dx \\ u'(0) = u'(1) = 0, \quad u \in C^2[0,1]. \end{cases} \tag{25}$$

where u denotes the polarization and E is a constant applied field.

The corresponding Euler-Lagrange equation is

$$\begin{cases} -\varepsilon^2 u''(x) + f'(u) = E \text{ in } [0,1], \\ u'(0) = u'(1) = 0, \quad u \in C^2[0,1]. \end{cases} \tag{26}$$

In order for (26) to have a solution, the following equality should hold

$$E = \int_0^1 f'(u)\, dx.$$

It is immediate that $u = \lambda$ is a solution for any $\lambda \in \mathbf{R}$. We then choose solutions satisfying $\int_0^1 u = \lambda$ for each $\lambda \in \mathbf{R}$. If u is one of such solutions, then u is a solution of

$$\begin{cases} -\varepsilon^2 u''(x) + f'(u) = \int_0^1 f'(u)\, dx \text{ in } [0,1], \\ u'(0) = u'(1) = 0, \quad \int_0^1 u\, dx = \lambda, \quad u \in C^2[0,1]. \end{cases} \tag{27}$$

The equation (27) is the Euler-Lagrange equation for the problem

$$\begin{cases} \mathbb{E}(u) = \int_0^1 \left\{ \frac{\varepsilon^2}{2}(u'(x))^2 + f(u) \right\} dx \\ \text{subject to} \\ u'(0) = u'(1) = 0, \quad \int_0^1 u\, dx = \lambda, \quad u \in C^2[0,1] \quad \lambda \in \mathbf{R}. \end{cases}$$

As a summary, we can rephrase the problem as:

Suppose that we have an antiferro or ferroelectric system governed by (25), and that the net polarization without electric fields may or

may not differ from λ. Find an electric field E and the corresponding polarization resulting in a net polarization being λ.

This is considered as a control problem which can be applied in reality. It is very physical in the sense that we don't restrict the solution space to $\int_0^1 u = \lambda$ at the beginning. It is worthwhile to mention that the problem addressed above is slightly different from the following problem

$$\begin{cases} \mathbb{E}(u) = \frac{1}{2}\int_0^1 \left\{\varepsilon^2(u'(x))^2 + f(u) - Eu\right\} dx, \\ u'(0) = u'(1) = 0, \quad \int_0^1 u = \lambda, \quad u \in C^2[0,1], \quad \lambda \in \mathbf{R}. \end{cases}$$

Furthermore, using the relation $E = \int_0^1 f'(u)\,dx$ we can obtain a hysteresis loop between u and E by varying the value of λ, where u is a solution of (27).

From the previous section, we obtained finitely many nontrivial solution branches bifurcating from the trivial solution $(0,\lambda) \in \mathcal{X} \times \mathbf{R}$. If (w,λ) is a solution on the curves, then $u = w + \lambda$ is a solution of (27) with the corresponding field $E = \int_0^1 f'(w(s) + \lambda)\,ds$. For each k in theorem 3.5, we can obtain a single or double loop corresponding to the nontrivial solution branch(es) for each k.

Theorem 4.1. *Let us assume all notations and results in theorem 3.5. For each k,*

1. *(ferroelectrics) there is one corresponding single hysteresis loop.*
2. *(antiferroelectrics with α)*
   - *(1) if $\alpha \in (-\frac{7}{5}, 1]$, then there is one corresponding double loop.*
   - *(2) if $\alpha \in (1,\infty)$, then there is a single hysteresis loop for $1 \le k \le k_0$ and a double loop when $k_0 < k \le K$.*

Furthermore, if $k < l$, then the hysteresis loop corresponding to l lies inside the one corresponding to k.

Proof. It suffices to prove that the closed nontrivial solution branch $\mathcal{C}^s_{k,\mathcal{F}}$ produces a hysteresis loop for ferroelectrics. For each $1 \le k \le M$, let $\mathcal{S}_k$ be the set of all trivial solutions $(0,\lambda)$ in $C^2[0,1]\times\mathbf{R}$ for $|\lambda| \ge \eta_k$. For $(\lambda,u) \in \mathcal{S}_k$, we get

$$u = \lambda, E = \int_0^1 \lambda(\lambda^2 - 1)\,dx = \lambda^3 - \lambda = u^3 - u.$$

Hence the set $\mathcal{S}_k$ corresponds to two curves in $E - u$ plane, which are the two connected components of

$$\{(E,u) : E = u^3 - u, |u| \ge \eta_k\}.$$

For $(\lambda, u) \in \mathcal{C}^s_{k,\mathcal{F}}$, we note that

$$(E, u) \to (\pm E_k, \pm\eta_k) \text{ as } \lambda \to \pm\eta_k,$$

where $E_k = \int_0^1 (\eta_k^3 - \eta_k)\, dx < 0$. This implies that $\mathcal{C}^s_{k,\mathcal{F}}$ represents a closed curve $\mathcal{H}_k$ in $E-u$ plane, which connects (E_k, η_k) to $(-E_k, -\eta_k)$ and vice versa. Moreover, the curve $\mathcal{H}_k$ doesn't include $(0, 0)$, and has a symmetric property that $(-E, -u) \in \mathcal{H}_k$ whenever $(E, u) \in \mathcal{H}_k$. This completes the proof. □

In Figures 3 - 5, we draw schematic shapes of hysteresis loops, which are corresponding to the nontrivial solution branches drawn in Figures 1 - 2. If $\alpha \le -\frac{7}{5}$, then no bifurcation occurs. This gives a loop in Figure 3(L). In Figures 3(R) - 5, the inner(outer) most closed loop (s) is (are) corresponding to the inner (outer) most nontrivial solution branch(es) $(-\lambda_k^1, \lambda_k^1)(\mathcal{C}^s_{k,\mathcal{F}})$ in Figures 1 - 2.

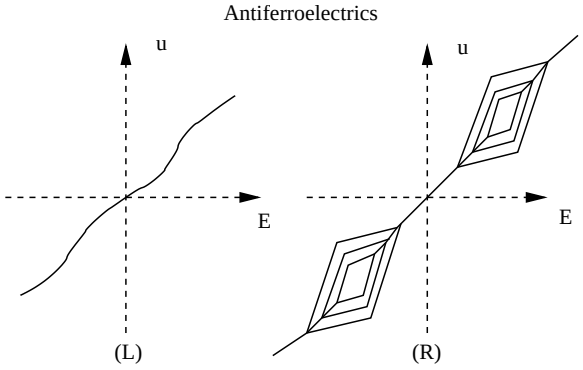

Figure 3: (L): $\alpha \le -\frac{7}{5}$, (R): $\alpha \in (-\frac{7}{5}, 1)$ is far away from 1

4.2 Applications

Example 1. Let us consider barium titanate, B_aTiO_3, known as a perfect ferroelectric solid. By the properties of the crystal in the cubic and tetragonal phase, the spontaneous polarization **P** is represented by $(0, 0, P)$ when an electric field is applied to the pseudo-cubic [001] direction [10, pp. 110]. This is an example of the problem (25).

The series of hysteresis loops reported in [14], which are obtained from experimental data at several different temperatures, can be produced from theorem 4.1 by varying α from α_1 to α_2 where $\alpha_1 < -\frac{7}{5}$ and $\alpha_2 > 1$ (see outer loops in Figures 3 - 5). This confirms that our analysis agree qualitatively with real experimental data.

We note that the coefficients a_{12}, a_{112}, a_{123} are not involved in the one dimensional model discussed in section 2. In fact, these constants

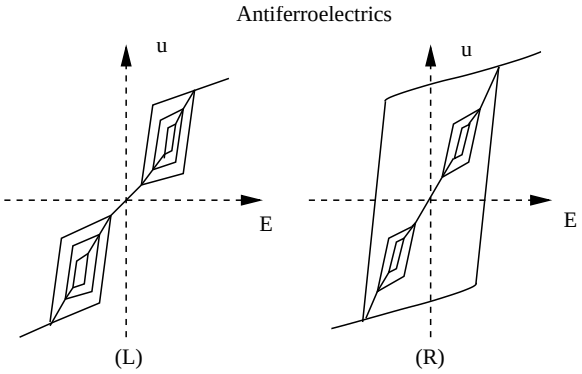

Figure 4: (L):$\alpha \in (-\frac{7}{5}, 1)$ is close to 1, (R): $\alpha(> 1)$ is close to 1

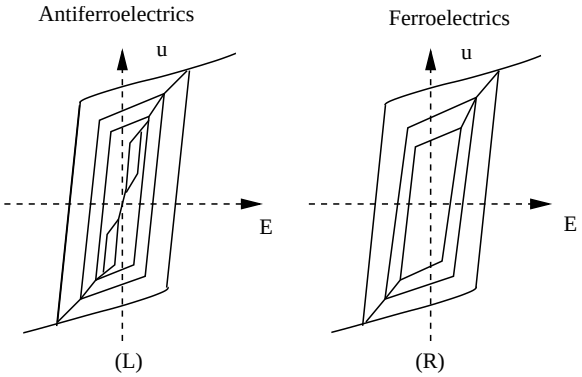

Figure 5: (L): $\alpha(> 1)$ is far away from 1, (R): Ferroelectrics

play an important role in the phase transition to orthorhombic and rhombohedral phases. For the transition to orthorhombic phase, let us apply an electric field along [110] direction and the polarization field $\mathbf{P}$ be represented by $\mathbf{P} = (P_1, P_2, 0)$ where $P_1 = P_2$. Denoting $P_1 = P_2 = \frac{1}{\sqrt{2}}u$, the polynomial in F_P can be simplified as

$$a_1 u^2 + \left(a_{11} + \frac{a_{12}}{4}\right) u^4 + \left(\frac{a_{111} + a_{112}}{4}\right) u^6.$$

Similarly, if the field is applied along [111] direction, then it induces a phase transition to a rhombohedral phase. So, if we let

$$\mathbf{P} = \left(\frac{u}{\sqrt{3}}, \frac{u}{\sqrt{3}}, \frac{u}{\sqrt{3}}\right),$$

then the polynomial is

$$a_1 u^2 + \left(a_{11} + \frac{a_{12}}{3}\right) u^4 + \left(\frac{a_{111} + a_{112}}{9}\right) u^6.$$

This also illustrates that shapes of hysteresis loops are depending on the directions of applied fields. Some of experimental data are found in [10, pp. 333].

Example 2. As another example, we consider the one dimensional problem discussed in section 2. From (5), we have

$$\frac{dv_1}{dx} = \frac{q_{12} P_0^2}{C_{11}} u^2 + \frac{s}{C_{11}}, \tag{28}$$

which gives a relationship between strain and the polarization. Having already studied hysteresis loops between polarization and an applied field, it is easy to get hysteresis loops associated with strain ($\frac{dv_1}{dx}$) and applied fields by (28). Experimental results in [19] show existence of different types of hysteresis loops depending on materials. Schematic drawings of these loops are depicted in Figure 6. We finally notice that a single $u - E$ loop generates a typical butterfly loop between strain and an applied field.

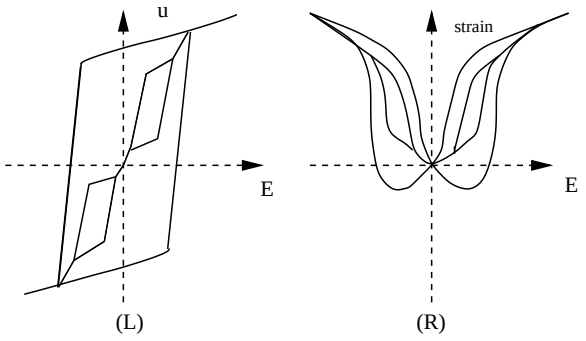

Figure 6: (R) describes corresponding loops to (L)

5 Conclusion

We studied Ginzburg-Landau types of the energies arising from modeling ferroelectric materials, and discussed the relationship between polarization and an applied field. We proved existence of finitely many branches of equilibrium configurations of the energies. By treating applied fields as Lagrange multipliers, we accomplished existence of

finitely many nested hysteresis loops which is a signature of finer structure of a ferroelectric material. We obtained critical fields which can induce nucleations during polarization reversal process. We also presented examples for applications confirming that current analysis explain physical phenomena occurring in the literature. In particular, new findings obtained from these sections are capable of visualizing solutions by numerical simulations and also providing ideas for modeling problems in material sciences, such as modeling of ferroelectric liquid crystals [18].

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