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Stochastic Dynamics of 180° domains

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Abstract

The goal of this work is to provide a simple and general framework for 180° domain analysis in ferroelectric thin films. In this work we construct mathematical and numerical description of complex ferroelectric systems and use it to advance our knowledge and understanding of 180° domains alongwith dynamic creep behavior.The main theoretical tool we have employed is an appropriate Landau-Ginzburg-Devonshire (LGD) model, and its time-dependent generalizations.Finite element computation is adopted during the expedition.[1]

Keywords— LGD model,Finite Element Analysis, Stochastic Interface Dynamics

1 Introduction

A ferroelectric material shows a large variety of applications in forms of actuators, sensors, nonvolatile random access memory systems, ultrasound transducers etc. Different rich technological aspects has led to extensive study of the material properties of ferroelectric materials. Domain evolution and domain movement are critically involved with such applications. This films of perovskites like $BaTiO_3$, $LiNbO_3$ are the materials in which there have been efforts to explore the behaviour of 180° and 90° domains. Although experimental measurements of domain growth rates in the well-established ferroelectrics $PbTiO_3$ and $BaTiO_3$ have been made, there is paucity of knowledge about domain dynamics. Several experiments show how a domain forms, nucleates and moves during electric switching [11, 9]. Some computational models have been developed to study domain formation and switching [6]. Collins et al have provided a theoretical stochastic model to study domain wall dynamics in ferrodistortive materials [6]. From a first principles density functional theory based study by Young et al have provided a method to study nucleation and growth mechanism, in connection with ferroelectric domain wall motion [6].

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In this article, epitaxial thin films (2D) in the context of finitely distributed elements is modeled. Time dependent Landau Ginzburg (TDGL) theory is the basis of our model development. The numerical scheme is presented in section 2. Results of finite element method are shown in section 3. In section 4, we demonstrate how domain wall moves and what is the dynamic behaviour of such moving domains. For this purposes, we begin with a simplistic but effective method of finite difference in time domains (FDTD). Experimental resemblance and further studies with results of FDTD method are described in section 5. Section 6 & 7 contain discussion and summary of our entire work.

2 TDLG Model

Nucleation and growth of the domains drive polarization reversal in ferroelectrics. Sideways expansion of the domains happens when an external electric field is applied to a ferroelectric. We use a simplified model to describe the main characteristic features of this sideways motion. The approach involves the use of the time-dependent Ginzburg-Landau theory. The model assumes that the interfacial and domain dynamics are completely determined by the evolution of the order parameter.

The Landau-Ginzburg (LG) theory includes significant spatial variations of the polarization within the phenomenological Landau-Devonshire model of free energy[13]. Slow variations in the direction of the polarization lead to an additional contribution to the free energy density (ϕ) which is proportional to $|\Delta P|^2$. We restrict our study to the case of a single-component order parameter, namely the polarization. We note that the spatial varying term also preserves the symmetry. Hence the Landau-Ginzburg free energy is [1, 3, 10]

$$\phi = -\frac{1}{2}AP^2 + \frac{1}{4}BP^4 + \frac{\delta}{2}\{\nabla P\}^2 - EP, \tag{1}$$

where $A = A_0(T - T_c)$, where A_0, B, δ are positive constants and T_c is the phase transition temperature in the absence of the applied electric field. The parameter E is the external electric field which plays a crucial role for domain dynamics. Evolution of domain dynamics will be determined by spatio-temporal variation of the order parameter polarization by the following kinetic equation

$$\frac{\partial \mathbf{P}}{\partial t} = -\Gamma \frac{\partial \phi}{\partial \mathbf{P}}. \tag{2}$$

Hence the time-dependent Landau-Ginzburg (TDLG) equation is (using Eq.(2.1) and Eq(2.2)),

$$\frac{\partial \mathbf{P}}{\partial t} = \Gamma\delta\nabla^2 P + \Gamma(AP - BP^3 + E). \tag{3}$$

The problem posed by Eq.(2.3) is nonlinear and therefore difficult to solve analytically. That is why we solve the problem numerically. The usual approach is to discretize the problem in space by some finite elements and then watch what is the transient response. The meaning of the latter is amplified below.

3 Finite Element Modelling

For studying macroscopic domain dynamics of ferroelectrics, we have persued a finite element approach to solve the TDLG equations. The equations can be solved analytically some extent [6] in one dimension, but boundary condition implementation and exactness are still beyond our scope. The finite element method is most suitable for structural studies. The basic building blocks of finite element approach are

1. Continuum is discretized in a finite number of elements of geometrically simple shapes.
2. Finite number of nodes will connect such elements.
3. Dislocation of the nodes will provide our expected variables.

4 Simulation Details

4.1 Space discretization

The structure of ferroelectric thin films are divided into triangular finite elements [Fig.1]. N number of such elements contribute the whole spatial structure in x-y plane where $N = 2(m-1) \times (n-1)$, m and n are number of nodes along x and y axes respectively. The polarization is interpolated by a linear shape function inside the element with weighted residuals [4].

4.2 Weak Formulation

We seek an approximate solution $u(x, y, t)$ for the unknown function $\mathbf{u}(x, y, t)$ in terms of a series involving basis functions (or, trial functions) with unknown coefficients. The requirement on the choice of basis functions is that they chose set of functions should be linearly independent and should represent a complete family of basis functions and can be expressed as a linear combination of these basis functions. Let us assume an approximate solution $u(x, y)$ as:

$$u(x,y) \approx u_0(x,y) + \sum_j C_j g_j(x,y) \tag{4}$$

where, $u_0(x)$ is a function which satisfies the non-homogeneous form of essential boundary conditions. $g_j(x, y)$; j = 1, 2, 3,... represent a family of basis functions and are chosen so as to satisfy the homogeneous essential boundary conditions of the problem. Applying weighted residual method, we formulate the PDE in a weak solution with weight $W(x, y)$

$$\int_\Omega w_i \left[\frac{\partial u}{\partial t} - \Gamma(K\nabla^2 u - Au + Bu^3 - E) \right] d\Omega = 0 \tag{5}$$

where, w_i is arbitrary weighting functions. Apply Green function to the quadratic term

$$\begin{aligned} \int_\Omega w_i \left[\Gamma(K\nabla^2 u) \right] d\Omega &= \Gamma K [\int_\Omega \begin{bmatrix} \frac{\partial w_i}{\partial x} \\ \frac{\partial w_i}{\partial y} \end{bmatrix} \begin{bmatrix} \frac{\partial u}{\partial x} & \frac{\partial u}{\partial y} \end{bmatrix} d\Omega \\ & \quad - \int_L w_i \nabla u \cdot \mathbf{n} dL] \end{aligned} \tag{6}$$

In Galerkin's weighted residual method, the weighting functions are identical to the trial functions, i.e.

$$w_i = h_i, \qquad i = 1, 2, 3 \tag{7}$$

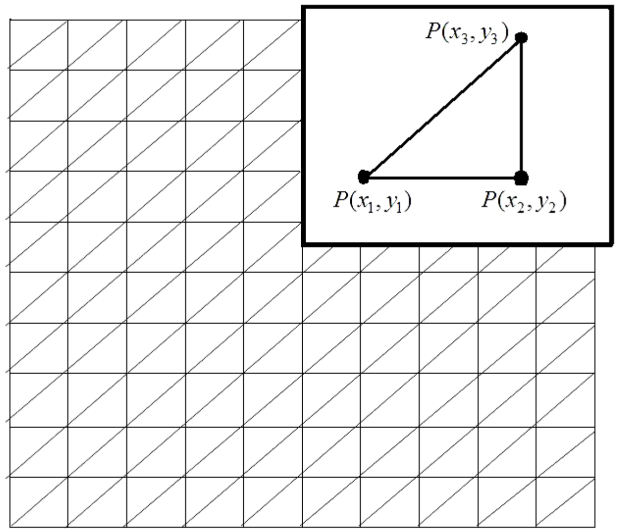

Figure 1: Element mesh (Inset: A triangular element used for calculating polarization)

4.3 Shape function

Fig.(2) depicts a surface of ferroelectric material The 2D volume represents the domain of a boundary value problem to be solved. For simplicity, at this point, we assume a two-dimensional case with a single field variable $P(x, y)$ to be determined at every point x, y such that a known TDLG equation is satisfied exactly at every such point or every node. Following this way, we will arrive at an exact mathematical solution .A small triangular element that encloses a finite-sized sub domain of the area of interest is shown in inset of Fig.(2.1). We are treating the problem as a two-dimensional. The vertices of the triangular element (nodes) are specific points in the finite element at which the value of the field variable is to be explicitly executed. For the three-node triangle, the field variable is described by the approximate relation

$$u(x, y) = h_1(x, y)u_1 + h_2(x, y)u_2 + h_3(x, y)u_3, \tag{8}$$

where $u_1, u_2\&u_3$ are the values of the field at three nodes and $h_1, h_2\&h_3$ are shape functions. Now we have to evaluate the nodal values taking them as constant. We express the field variable in the polynomial form

$$u(x, y) = a_0 + a_1x + a_2y \tag{9}$$

Then for the three nodes 1 2 and 3

$$\begin{bmatrix} 1 & x_1 & y_1 \\ 1 & x_2 & y_2 \\ 1 & x_3 & y_3 \end{bmatrix} \begin{bmatrix} a_0 \\ a_1 \\ a_2 \end{bmatrix} = \begin{bmatrix} u_1 \\ u_2 \\ u_3 \end{bmatrix} \tag{10}$$

Under inversion

$$
\begin{aligned}
a_0 &= \frac{1}{2A}[u_1(x_2y_3 - x_3y_2) + u_2(x_3y_1 - x_1y_3) \\
&\quad + u_3(x_1y_2 - x_2y_1)] && (11) \\
a_1 &= \frac{1}{2A}[u_1(y_2 - y_3) + u_2(y_3 - y_1) \\
&\quad + u_3(y_1 - y_2)] && (12) \\
a_2 &= \frac{1}{2A}[u_1(x_3 - x_2) + u_2(x_1 - x_3) \\
&\quad + u_3(x_2 - x_1)] && (13)
\end{aligned}
$$

where A is the area of the triangular element determined by

$$
A = \frac{1}{2}det \begin{vmatrix} 1 & x_1 & y_1 \\ 1 & x_2 & y_2 \\ 1 & x_3 & y_3 \end{vmatrix} \tag{14}
$$

using Eq.(2.8-2.14) and after a rearrangement, we get

$$
\begin{aligned}
u(x,y) &= \frac{1}{2A}\{[(x_2y_3 - x_3y_2) + (y_2 - y_3)x + (x_3 - x_2)y]u_1 && (15) \\
&\quad + [(x_3y_1 - x_1y_3) + (y_3 - y_1)x + (x_1 - x_3)y]u_2 && (16) \\
&\quad + [(x_1y_2 - x_2y_1) + (y_1 - y_2)x + (x_2 - x_1)y]u_3\} && (17) \\
h_1(x,y) &= \frac{1}{2A}[(x_2y_3 - x_3y_2) + (y_2 - y_3)x + (x_3 - x_2)y] && (18) \\
h_2(x,y) &= \frac{1}{2A}[(x_3y_1 - x_1y_3) + (y_3 - y_1)x + (x_1 - x_3)y] && (19) \\
h_3(x,y) &= \frac{1}{2A}[(x_1y_2 - x_2y_1) + (y_1 - y_2)x + (x_2 - x_1)y] && (20)
\end{aligned}
$$

4.4 FE formulation

To develop the finite element equations, a two-node linear element for which

$$
u(x,y) = h_1(x,y)u_1 + h_2(x,y)u_2 + h_3(x,y)u_3, \tag{21}
$$

is used with Galerkin's method. For Eq.(2.5), the residual equations are expressed as

$$
\int_{x_1}^{x_2}\int_{y_1}^{y_2}\left[\frac{\partial P}{\partial t} - \Gamma(K\nabla^2 P - AP + BP^3 - E)\right]h_i(x,y)dxdy = 0 \qquad i = 1,2,3 \tag{22}
$$

Time domain simulation is based on forward interpolation. Nonlinearity approximated with

$$
u^3 \approx (h_i)^2 u \tag{23}
$$

therefore the whole process become quasi-linear. As far as stability of concern, quasi-linear solution approximates the nonlinear part. Thus we get a numerical solution based on theoretical finite element basis where we have approximated the solution and in a self-organizing manner[12, 14]. Whole process has chosen such solutions that converges the equation. Fig.(2), we have shown how periodic domain evolves and how electric field switches it respectively.

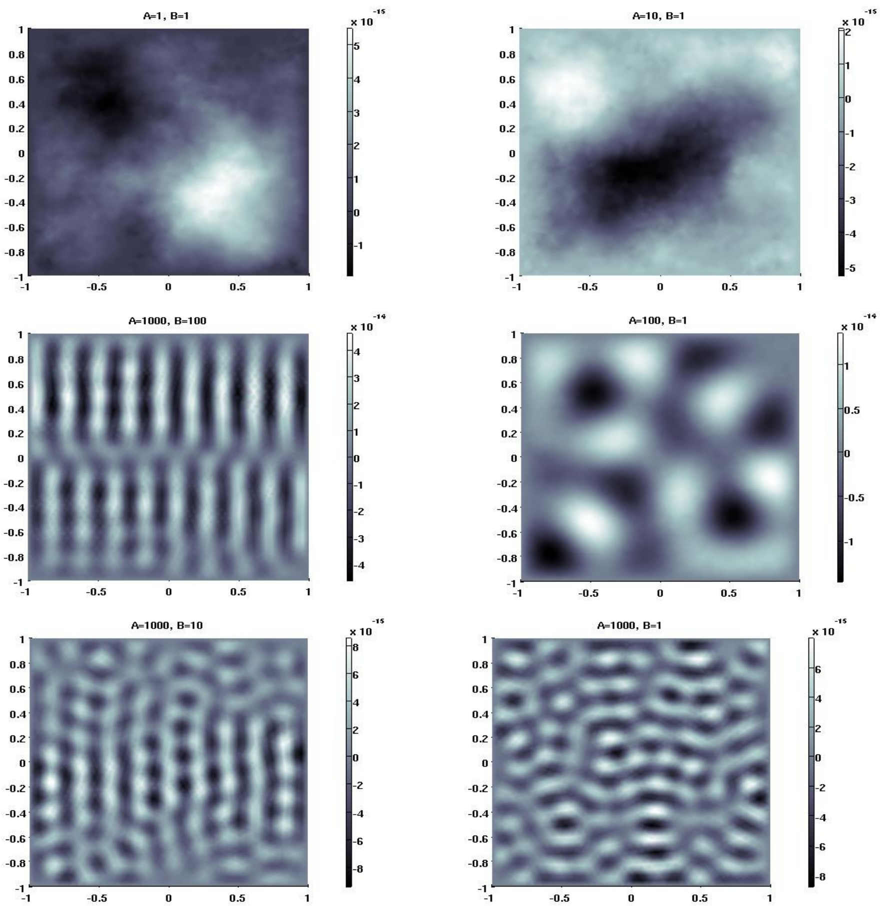

Figure 2: Topographic appearances of 180° domains at different time steps.

5 Analytical Approach

An approximate solution of nonlinear partial differential equation like (TDLG)

$$\frac{\partial P}{\partial t} = \delta \frac{\partial^2 P}{\partial x^2} + (AP - BP^3 + E) \tag{24}$$

can be given by [7]

$$P(x,t) = \frac{\beta}{F}\frac{\partial F}{\partial x} + \lambda \tag{25}$$

where $\beta = \pm\sqrt{\frac{2\delta}{b}}$ and $\lambda =$ any of the roots of the equation

$$-b\lambda^3 + a\lambda + E = 0 \tag{26}$$

we have taken the only real solution

$$\lambda = \frac{(\frac{2}{3})^{\frac{1}{3}} a}{(9b^2E + \sqrt{3}\sqrt{-4a^3b^3 + 27b^4E^2})^{\frac{1}{3}}} + \frac{(9b^2E + \sqrt{3}\sqrt{-4a^3b^3 + 27b^4E^2})^{\frac{1}{3}}}{2^{\frac{1}{3}}3^{\frac{2}{3}}b} \tag{27}$$

$F(x,t)$ depends on coefficients of Eq(2.23). and can be expressed as

$$F(x,t) = C_1 exp(k_+x + s_+t) + C_2 exp(k_-x + s_-t) + C_3 \tag{28}$$

where
$k_\pm = -\frac{1}{2} \pm \frac{1}{2}\sqrt{q_1^2 - 4q_2}$ and $s_\pm = -k_\pm^2 p_1 - k_\pm p_2$ with
$p_1 = -3\delta$, $q_1 = \frac{3b\beta\lambda}{2\delta}$, $p_2 = -3b\beta\lambda$, $q_2 = \frac{1}{2\delta}(3b\lambda^2 - a)$
Now if we consider a special case where $C_1 = C_2 = C$, $C_3 = 0$, $k_1 = -k_2 = k$, and $s_1 = -s_2 = s$ then

$$F(x,t) = C[exp(kx + st) + exp(-kx - st)] \tag{29}$$

$$P(x,t) = \frac{C\beta}{k} tanh(kx + st) \tag{30}$$

Therefore the velocity will be s (or -s).

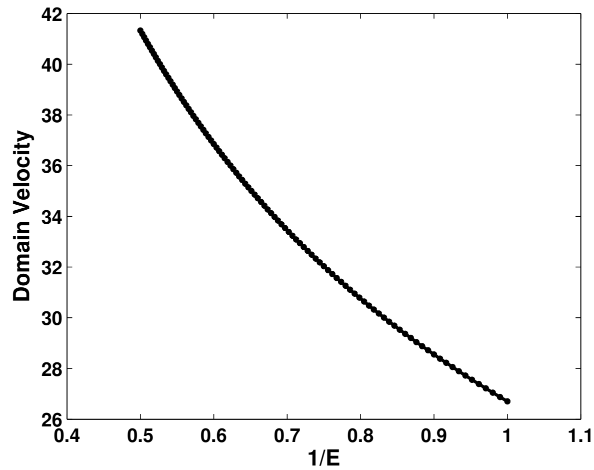

Figure 3: Domain wall velocity in terms of reciprocal of Electric field. a=1,b=1,$\delta = 1$

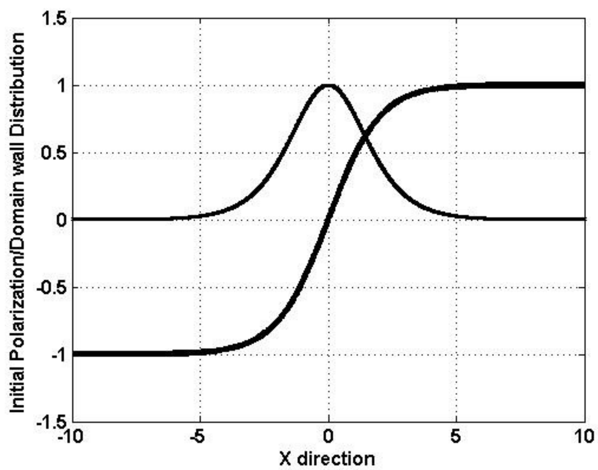

Figure 4: Initial domain decoration in space, domain and domain wall width

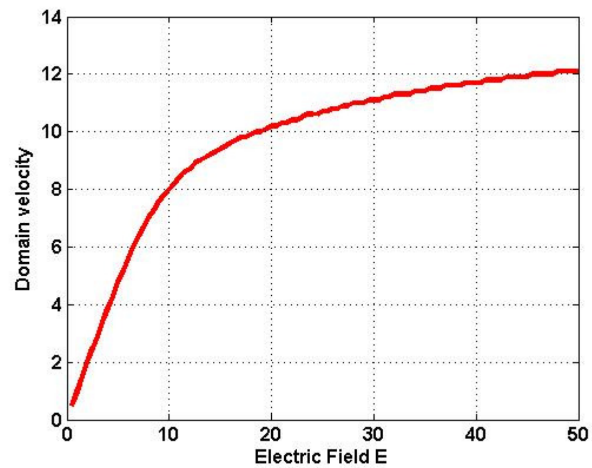

Figure 5: Domain wall velocity in terms of Electric field E by FDTD method

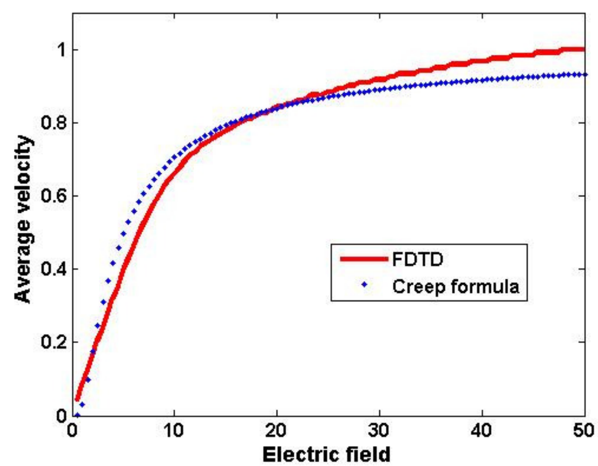

Figure 6: 180° domain wall showing creep behaviour

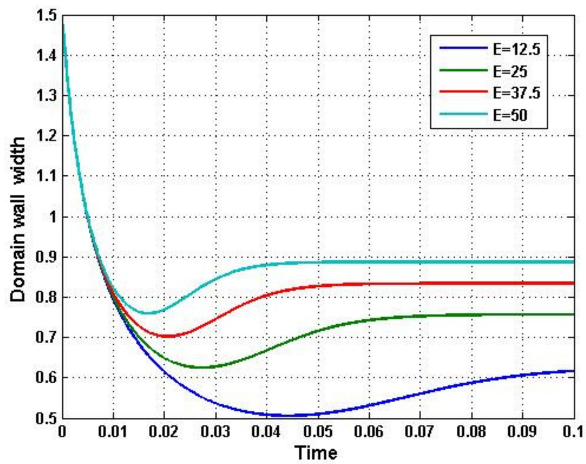

Figure 7: Domain wall width in time domain

In previous section we developed how ferroelectric domain evolves via TDLG equation through finite element method. Now we will be elaborating how these domain moves under electric field switching. We calculated velocity in one dimension domain array with 180° pattern. For this study, it is enough to solve TDLG equation in 1D.

$$\frac{\partial \mathbf{P}}{\partial t} = \Gamma\delta\frac{\partial^2 P}{\partial x^2} + \Gamma(AP - BP^3 + E) \tag{31}$$

is

$$P(t=0,x) = P_0 tanh\left(\frac{x}{r_c}\right) \tag{32}$$

To solve TDLG equation, we used centered difference approximated finite difference scheme for spatial sampling whereas for time sampling, we used forward approximation.

$$\frac{\partial P(t,x)}{\partial t} \approx \frac{P(t+\delta t,x) - P(t,x)}{\delta t} \tag{33}$$

$$\frac{\partial^2 P(t,x)}{\partial x^2} \approx \frac{P(t,x+\delta x) - 2y(t,x) + y(t,x-\delta x)}{\delta x^2} \tag{34}$$

The Ginzburg term $\frac{\partial^2 P}{\partial x^2}$ in this equation requires left as well as right boundary condition simultaneously keeping in mind that the physical domain structure is 180°. Therefore our boundary conditions are

$$P(t=0,x) = P_0 tanh(\frac{x}{r_c}) \text{ [Initial condition]} \tag{35}$$

$$P(t,x=-L) = -1, \qquad P(t,x=L) = 1; \tag{36}$$

therefore initial domain wall distribution is

$$D_w = \frac{\partial P}{\partial x}(t=0,x) = \frac{P_0}{r_c} sech^2 \frac{x}{r_c}. \tag{37}$$

Velocity is calculated taking maximum value of $\frac{\partial P}{\partial x}$ as reference point . Domain wall width is taken as standard deviation σ calculated by Full width at half maximum and $FWHM = 2\sqrt{2ln2}\sigma$. Other parameters $T = 18$; $T_c = 24$; A1=0.0410; A=A1*(T-Tc); B=100;

6 Fluctuations in Ferroelectrics

How does the system with some fluctuations approach towards equilibrium ? Earlier, we have discussed L-K equation with fluctuations. The L-K equation with thermal fluctuations is nothing but Langevin equation which is a stochastic differential equation.

$$\frac{dx}{dt} = -\Gamma\frac{\partial\phi}{\partial x} + \eta(t) \tag{38}$$

where Γ the Kinetic coefficient, $\eta(t)$ is the random or fluctuating force. The distribution of $\eta(t)$ is Gaussian, and its correlation function obeys

$$< \eta(P,t)\eta(P',t') >= 2\Gamma k_B T\delta_{x,x'}\delta(t-t') \tag{39}$$

Corresponding fokker planck equation will be

$$\frac{\partial f(P,t)}{\partial t} = \frac{\partial}{\partial P}\left(f(P.t)\frac{\partial\phi}{\partial P}\right) + \Gamma\frac{\partial^2 f(P,t)}{\partial P^2} \tag{40}$$

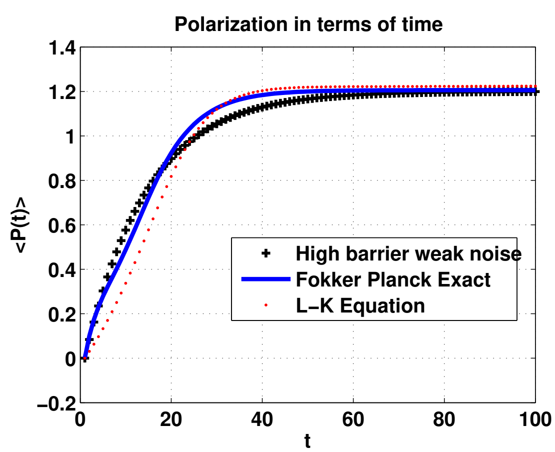

Figure 8: Numerically simulated result (P(t) vs t) from Landau-Khalatnikov equation, Fokker Planck equation and Kramer's Treatment of high barrier weak noise.

For this case the free energy $\phi = -a_0(T-T_c)P^2 + bP^4 - EP$. The equilibrium probability is obtained if we equate $\frac{\partial f}{\partial t} = 0$, hence

$$f^{eq}(P) = \frac{exp[-\phi(P)/k_B T]}{\int dP' exp[-\phi(P')/k_B T]]} \tag{41}$$

7 Results and Discussion

In conclusion, lateral domain wall motion in ferroelectric thin films is a creep process. Domain wall motion is governed under influence of external electric field (Fig.(5))[8, 6, 11, 12].Fig.4 shows width of typical an 180^0 domain. The velocity data fits well with creep formula as described by Tybell et al [11] (Fig.(4))

$$v = Ce^{\alpha E^{\mu}} \tag{42}$$

α depends on A, Creep dynamical exponent can be fitted as $\mu \approx 1$. Our results suggest a stable and general method for measuring velocity and wall thickness of ferroelectric domains.

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