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Monte Carlo investigation of hysteresis properties in ferroelectric thin-films under the effect of uniaxial stresses

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Abstract

The uniaxial stress dependence of the hysteresis behavior of ferroelectric films was studied. The DIFFOUR model was modified to include the uniaxial stress effect. Both the uniaxial stress and the external electric field were applied on the out-of-plane direction of the films. The polarization was measured with varying the magnitude of the applied stress and the electric field frequency via the dynamics of the polarization reversal in terms of hysteresis. The study was taken by means of Monte Carlo simulations using the spin-flip Metropolis algorithm. From the results, the district dependence of hysteresis behavior on frequency between low frequency and high frequency was prominent. On the other hand, the remanent and the coercivity significantly decreased with increasing applied stresses. Moreover, the areas under the hysteresis loops also decreased indicating smaller magnitude of energy dissipation. The results agree well with related experiments where applicable.

Keywords: Ferroelectric thin-films; Monte Carlo; Uniaxial stress; Hysteresis

1. Introduction

Ferroelectric thin-films have recently been of wide interest in view of both technological and fundamental importance [1,2]. Of a particular interest is the technological applicability such as high-speed ferroelectric recording media in which high areal densities and high reliability are in demand [1]. Therefore, it is necessary to understand the response of ferroelectric domain switching to electric field corresponding to specific material structures in detail. Nevertheless, for the sake of simplicity, theoretical studies on ferroelectric multi-layers are usually performed on an ideal stress-free system. However, real materials used in many applications are often affected by crystalline anisotropy caused by external mechanical stress, or internal strain induced by misfit in lattice spacing at the interfaces between ferroelectric layers and the substrate. Furthermore, ferroelectric thin-films under stresses were found to have their polarization behavior altered leading to substantial changes in phase transition between ferroelectric and para-electric phases [3,4]. Consequently, it is very important to include the applied stress to model real materials.

In this work, the uniaxial stress dependence of the ferroelectric dynamic properties in thin-films was studied. To outline, the study was firstly done by proposing the DIFFOUR Hamiltonian that includes the uniaxial stress effect. Then, by means of Monte Carlo simulations, the polarization along the out-of-plane direction is investigated with varying the field frequency and uniaxial stress via the dynamics of the hysteresis. Finally, all the descriptions to these results are given in detail.

2. Methodologies

2.1. Spin Hamiltonian

In this study, the DIFFOUR Hamiltonian [5–7]

$$H=\sum_i\left(\frac{P_0^2}{2m}-\frac{a}{2}u_i^2+\frac{b}{2}u_i^4\right)-\sum_{<ij>}U_{ij}\vec{u}_i\cdot\vec{u}_j-E(t)\sum_i u_{iz} \quad (1)$$

was considered where $\vec{u}_i$ is the ferroelectric dipole spin at site i, $P_0^2/2m$ is the kinetic energy, a and b are the double-well

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potential parameters for the ferroelectric spins, and U_{ij} is the ferroelectric interaction. However, in this study, only the case that $\vec{u}_i$ is constant in magnitude was considered to underline the dynamics of the spin orientation in response to the field. Therefore, by proposing appropriate reference energy and introducing the stress effect, the Hamiltonian can be rewritten as

$$H = \sum_{<ij>} U_{ij}(\Delta l)\hat{u}_i \cdot \hat{u}_j - E(t)\sum_i u_{iz}, \tag{2}$$

where $\hat{u}_i$ is a unit vector referring to one of the possible 14 ferroelectric spin directions (6 from tetragonal and 8 from rhombohedral structures), $<ij>$ represents summation over the nearest pairs, and u_{iz} is the spin's z component. $E(t) = E_0 \sin(2\pi ft)$ is the electric field acting only on the out-of-plane direction of the films, where f and E_0 refer to frequency and amplitude respectively. Helical and free-boundary conditions were used for the in-plane (xy) and the out-of-plane (z) directions. In this picture, the magnitude of $\hat{u}_i$ is dimensionless, so both U_{ij} and E have a unit of energy. $U_{ij}(\Delta l)$ is a function of lattice distortion Δl, arising from the applied stress, and assumed to take a Lennard–Jones potential-like [8], i.e.

$$U_{ij}(r_{ij}) = U_0\left[\left(\frac{r_0}{r_{ij}}\right)^{12} - 2\left(\frac{r_0}{r_{ij}}\right)^{6}\right]. \tag{3}$$

Here, r_0 is the lattice spacing at a specific thermal equilibrium, U_0 is the ferroelectric interaction associated to r_0 ($\Delta l = 0$), and r_{ij} is the distance between site i and j. For zero stress (strain), $r_{ij} = r_0$ and $U_{ij} = -U_0$ so the system prefers ferroelectric phase. Since the Young's modulus is defined as $Y \equiv P/((r_{ij} - r_0)/r_0)$ where P is the stress (pressure), it is possible to write $r_{ij}/r_0 = 1 - (P/Y)$ and Eq. (3) as $U^z_{ij} = U_0[(1-(P/Y))^{-12} - 2(1-(P/Y))^{-6}]$. However, along the xy direction, there also exists the lattice distortion caused by the stress along the z direction. The ratio of the distortions between these two directions is defined as the Poisson ratio $\varepsilon \equiv -\Delta r^{xy}/\Delta r^z$ (where for many systems ranging from metal to ceramics, $\varepsilon \approx 0.3$ [9]). As a result, it is possible to write $\varepsilon = -\Delta r^{xy}/\Delta r^z = -(r^{xy}_{ij} - r_0)/(r^z_{ij} - r_0) = ((1 - r^{xy}_{ij})/r_0)/(r^z_{ij}/(r_0 - 1))$ which gives $r^{xy}_{ij}/r_0 = 1 - \varepsilon((r^z_{ij}/r_0) - 1) = 1 + \varepsilon(P/Y)$. Consequently, the ferroelectric interaction along the xy direction is $U^{xy}_{ij} = U_0[(1+(\varepsilon P/Y))^{-12} - 2(1+(\varepsilon P/Y))^{-6}]$. As a result, the Hamiltonian can be written as

$$H = \sum_{<ij> \in \text{in-plane}} U^{xy}_{ij}\hat{u}_i\hat{u}_j + \sum_{<ij> \in \text{out-of-plane}} U^z_{ij}\hat{u}_i\hat{u}_j - E(t)\sum_i u_{iz}. \tag{4}$$

2.2. Monte Carlo simulation

Throughout this study, U_0 was set as 1, so this re-defines the unit of temperature T as J/k_B (where k_B is the Boltzmann's constant), and electric field amplitude E_0 as a unit of U_0. The simulations were done at a temperature in the ferroelectric phase (where there exists hysteresis loops), i.e. $T = 1.0\ J/k_B$. The ratio P/Y was varied from 0.00 to 0.16. The simulation was done on bi-layer ferroelectric films where each single layer consists of ferroelectric unit cells connecting along the in-plane direction. The ferroelectric spins are assumed to reside in the unit cells and the system consists of $N = L \times L \times 2$ spins where $L \times L$ refers to number of spins in one monolayer. To minimize finite size effect, large L is required so in this study $L = 40$ was chosen. Trial simulations for larger sizes were also performed and it is found that for the range of parameters used in this study, the difference in hysteresis behavior is not significant. The unit time step was defined from one full simulation update of all sites of the lattice, i.e. 1 Monte Carlo step per site (mcs). The field frequency was varied from 0.001 to 4 mcs^{-1} and the field amplitude is fixed at $E_0/U_0 = 4$.

With the Hamiltonian proposed in Eq. (3), each system was assigned an initial random configuration and later on was passed to the thermal Monte Carlo updates, using the Metropolis algorithm [10]. In updating the system, each spin is assigned a new random direction, and the probability of accepting that new direction is proportional to

$$\text{probability} = \exp\left(-\frac{\Delta H}{k_B T}\right) \tag{5}$$

where ΔH is the energy differences between of the original and the new updating state. If the energy difference is less than zero, or a uniform random number in the range [0,1) is less than the probability given in Eq. (5), the new direction is accepted and the system is successfully updated, or else the considered spin is left untouched. The whole procedure is repeated until the simulation ends.

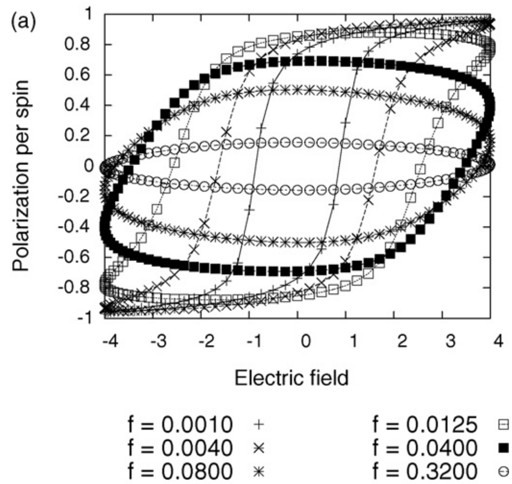
In measuring, with varying the uniaxial stress and the applied field frequency, each simulation waited for a few cycles to obtain steady hysteresis loops, and then the polarization per spin along the z direction was calculated, i.e.

$$p_r = \frac{1}{N}\left[\sum_i u_{iz}\right]. \tag{6}$$

Then, 1000 steady hysteresis loops were used to calculate average hysteresis loop for each condition.

3. Results and discussion

From the simulation results, with varying field frequency and stress, significant changes to the hysteresis loop were found. Fig. 1a shows the patterns for hysteresis loops for frequencies ranging from 0.001 to 0.320 mcs^{-1}. As can be seen, 2 distinct behaviors can be found for low frequency region, e.g. $f \leq 0.125\ \text{mcs}^{-1}$ and for high frequency region, e.g. $f > 0.125\ \text{mcs}^{-1}$. At low frequencies, the loops get bigger with increasing frequency. This is because, at low frequency, the field period is high and hence the sweeping time of the field per one hysteresis cycle is large. Ferroelectric spins then have time to follow the field leading to low phase-lag between the polarization and field and hence to a small hysteresis loop.



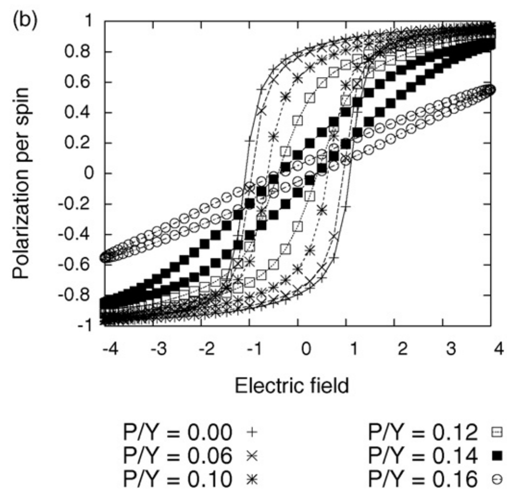


Fig. 1. Hysteresis loops of the bi-layered films (a) at zero-stress but varying frequency from $f = 0.0010$ to $f = 0.3200$ mcs^{-1}, and (b) at $f = 0.0010$ mcs^{-1} but varying stress from $P/Y = 0.00$ to $P/Y = 0.16$.

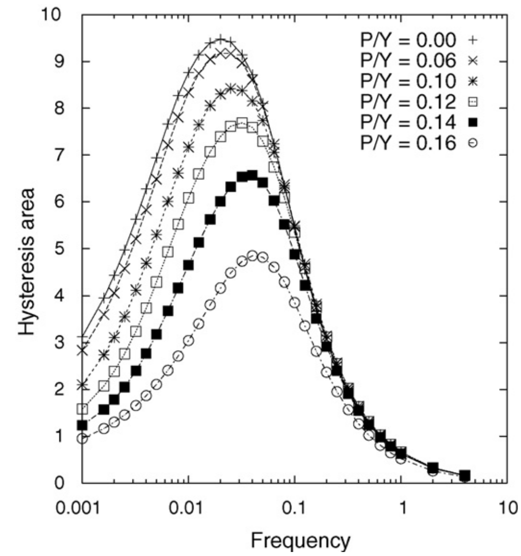


Fig. 2. Stress dependent of the hysteresis areas (arbitrary unit) of the bi-layered films as a function of the electric field frequency (mcs^{-1} unit) with varying stresses from $P/Y = 0.00$ to $P/Y = 0.16$.

However, with increasing the frequency in the low frequency region, the phase lag gets bigger and hence the hysteresis loop becomes larger. However, if the frequency is still increased, the loop reaches a maximum size at a certain frequency. Beyond this point, the frequency is very high which limits the dynamics of the spins and results in a smaller loop with an oval shape-like.

However, with a non-zero stress, e.g. in Fig. 1b, the decrease of the remanent p_r, the coercivity E_C, and the hysteresis loop-area were found. The reason is that the stress causes the z direction to become a 'hard axis'. As a result, the spins prefer to align in the xy direction or have their z-components align in an anti-parallel pattern to lower the energy. Consequently, both p_r and E_C reduce. In comparison with experiments, qualitatively, the decrease of the coercivity and the loop-area with increasing stress has trends that agree reasonably well with an experiment on ferroelectric material [11].

On the other hand, Fig. 2 shows the hysteresis loop area as a function of frequency for various stresses. The loop area increases for low frequency region and decreases for high frequency region with increasing frequency. At a particular frequency, the loop area is smaller for larger stresses. This is because the z direction is the stress induced 'hard axis'. In this way, the spins are loosely coupled along the z direction and hence it will be easier for the spins to catch up with the field so the inclined oval loop occurs with small loop area. However, with increasing the frequency at low frequencies, the phase lag gets larger but with higher stresses it will require higher frequency to reach the loop-area maximum point. This is why the frequency at the maximum area shifts to higher frequency for larger stress. Notice that the maximum area is smaller for larger stress because of smaller p_r and E_C.

4. Conclusions

In this study, Monte Carlo simulations were performed to study ferroelectric thin-films under the influence of uniaxial stress on ferroelectric hysteresis properties. The objective is to investigate the behavior of the spin-reversal along the out-of-plane direction with changes in the magnitude of the stress and the electric field frequency. With increasing frequency, it is found that the hysteresis area increases for low frequency and decreases for high frequency. On the other hand, the applied stress reduces p_r, E_C and the hysteresis area. This is due the stress causing the out-of-plane direction to be a 'hard axis' and the hysteresis results in smaller loop-area. The results qualitatively agree well with experiments where applicable.

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