Vol 18 No 4, April 2009
1674-1056/2009/18(04)/1665-04
Chinese Physics B
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Potts–Ising model for simulation of polarization switching in polycrystalline ferroelectrics*

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(Received 27 November 2007; revised manuscript received 23 October 2008)

This paper proposes a scheme based on the Potts and Ising models for simulating polarization switching of polycrystalline ferroelectrics using the Monte Carlo method. The polycrystalline texture with different average grain size is produced from the Potts model. Then Ising model is implemented in the polycrystalline texture to produce the domain pattern and hysteresis loop. The domain patterns and hysteresis loops have been obtained for polycrystalline texture with different average grain size. From the results of domain pattern evolution process under an applied electric field using this scheme, an extended domain, which covers more than one grain with polarization aligned roughly in the same direction, has been observed during the polarization reversal. This scheme can well reproduce the basic properties of polycrystalline ferroelectrics and is a valuable tool for exploring the physical properties of polycrystalline ferroelectrics.

Keywords: grain growth, ferroelectric properties, Potts–Ising model
PACC: 7780, 7780D, 7510H

1. Introduction

Polycrystalline ferroelectrics is of considerable interest due to their applications in electronic, microelectronic and electro-optical devices.[1−5] In these applications, the switching behaviour of ferroelectrics under an applied electric field is exploited such as in the design of nonvolatile random access memories. It has also been observed that the domain structure plays a significant role in determining the switching behaviour in these materials[6] Given that switching is the source of nonlinear electric displacement response for polycrystalline ferroelectrics, it should be possible to model hysteresis loops and domain structure obtained experimentally. Many theoretical works[7−9] have been already carried out based upon models of perfect single crystalline structure for ferroelectrics. Hwang *et al* [7] have developed a polarization switching model, which assumed that a single ferroelectric crystallite in a ceramic, to predict the electric displacement and strain hysteresis loops for the PLZT under varying electric fields. A phase-field model[9] has been developed for predicting the polarization switching and domain structure evolution under an applied electric field in ferroelectric polycrystals. The model takes into account the realistic grain structures as well as various energetic contributions, including elastic energy, electrostatic energy, and domain wall energy. A correlation between the domain structures in neighbouring grains was observed, and polarization switching in one grain was found to affect the switching in neighbouring grains.

In this paper, we develop a scheme that can simulate the basic properties of polycrystalline ferroelectrics from combination of the Potts model and the Ising model. The basic idea is that first the grain evolution process is simulated from the Potts model where the polycrystalline structure with different average grain size can be produced, then the Ising model is implemented in the polycrystalline texture to produce domain pattern and hysteresis loop. The remainder of this paper is organized as follows. First we explain the Potts–Ising model we have applied and show that it is able to equilibrate the system. The choices of the parameters for simulation are given in this section. In the next part, we show the hysteresis loops and the domain evolution during the polarization reversal as example to demonstrate the validity of this scheme. In the last section, we summarize our results.

*Project supported by National Natural Science Foundation of China (Grant No 10474057).
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2. Model and method

The Hamiltonian of a q-state Potts model is

$$H = -\sum_{\langle ij \rangle} \delta_{\sigma_i \sigma_j}, \tag{1}$$

where $\sigma_i = 0, 1, \ldots q-1$ are different spin states, but stands for the different grain orientations in this work, δ is the Kronecker operator, when $\sigma_i = \sigma_j$, $\delta = 1$, otherwise $\delta = 0$. The Potts model was first proposed by Potts as a generalization of the Ising model for simulating the critical transitions in magnetic materials with more than two degenerate states.[10] At an early stage, the Potts model was applied to simulate coarsening phenomena, namely grain growth.[11] Then it was modified to study many microstructural evolution problems including normal[12] and abnormal grain growth,[13] recrystallization,[14] etc. Since then, the Potts model has been widely used to simulate the grain growth in metal materials.[15,16] As the Monte Carlo method with Metropolis sampling usually produces a lower growth index and low simulation speed, different algorithms have been proposed to increase the simulation speed and improve the growth index.[15−18] Considering the reorientation attempts of the lattice sites that are limited to the orientations of the nearest neighbours, we obtain a growth index almost 0.5.[15] Another modification which allows taking into account alterations of the texture development is proposed. It is found that as the ratio of the mobility increases, the texture change occurs fast and the growth index is much near 0.5.[16] In order to better reflect the physical behaviour observed in nature, an object oriented approach is used to get the value of growth index 0.5 only at the first simulated stage.[17] Among them, minimum energy preferable orientation (MERO) algorithm has been proved to produce growth index very close to the theoretical value 0.5, while the simulation speed is much accelerated.[18] This method changes the node to optimum orientation corresponding to the system having the lowest energy. So in this work we use the MERO algorithm to produce the polycrystalline texture from Potts model. The grain size is controlled by the Monte Carlo steps (MCS). Therefore we can obtain polycrystalline texture with different average grain size.

After we have the polycrystalline texture with desired average grain size, spin-1/2 Ising model is implemented. The two spin states represent the two dipole orientation states in ferroelectrics. The Hamiltonian of the Ising model has the following form,

$$H = -\sum_{\langle ij \rangle} J_{ij} S_i^z S_j^z - 2\eta E(t) \sum_i S_i^z, \tag{2}$$

where $S_i^z = \pm 1$ stands two opposite directions of the dipoles, J_{ij} measures the interaction of pair interactions, here only the nearest-neighbouring couplings are taken into account in the simulation. The exchange strength $J_{ij} = J\cos(\theta_{ij})$, where θ_{ij} is the angle between orientation of σ_i and σ_j. When the two dipoles are within the same grain, we have $\sigma_i = \sigma_j$, otherwise $\sigma_i \neq \sigma_j$, that is the two dipoles lie in the grain boundary. The angle between the two grains is defined as $\theta_{ij} = 2\pi(\sigma_i - \sigma_j)/q$, η is the dipole moment, $E(t)$ is the applied external field. An electric field in sinusoidal form is applied to reverse polarization,

$$E(t) = E_0 \sin(\omega t), \tag{3}$$

where E_0 is the amplitude of the applied field, which is set to be larger than the coercive field in the simulation, ω is the frequency of the applied field, we set $\omega = 0.01$. The global polarization is taken the average of the dipole in the whole lattice, i.e.

$$P = \frac{\sum_i S_i^z \cos(\theta_i)}{N \times N}. \tag{4}$$

A two-dimensional square lattice of size 200×200 with periodic boundary conditions have been used for both simulations of grain texture and domain pattern. The MCS is used as time scale. The number of states in the Potts model is q=68, which represents the maximum number of grain orientation. Temperature T is set to 0.68 J, which is scaled by the Curie temperature of the two-dimensional infinite lattice, T_c=2.2692 J, so it is about $T = 0.3T_c$, below the Curie temperature.

3. Results and discussion

Figure 1 shows the polycrystalline textures with different average grain size and their corresponding domain patterns. In this work, we assume that the polycrystalline texture is static when the grains stopped grow, i.e., it does not evolve with time when the Ising model is implemented in the polycrystalline texture to simulate their ferroelectric properties. The average grain area is defined as the total lattice site 200×200 divided by the total number of the grains. So the unit of grain area is the number of lattice sites. The average grain size is the square root of the average grain area. To visualize the simulation results, the

orientation domain is presented by different grey scale due to different grain orientations. The dark stands for one direction of polarization, and the white represents the opposite direction. The domain patterns are obtained under an electric field at the saturated polarization. Since an electric field has been applied, every grain is in a single domain state.

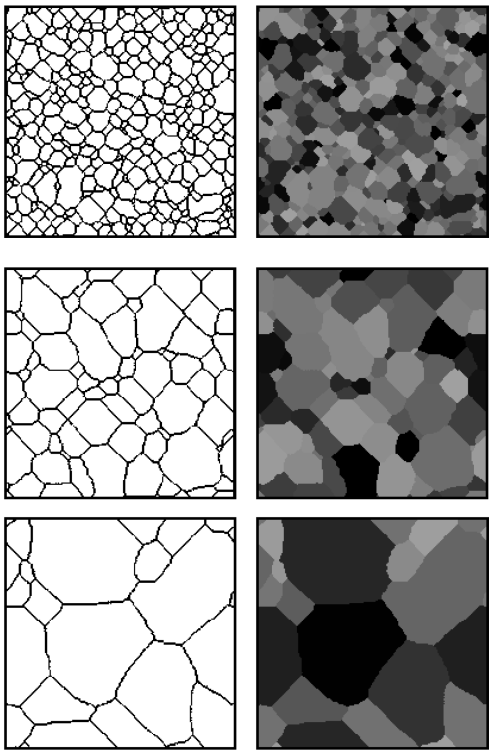


Fig.1. Grain textures with different average grain sizes from Potts model and their corresponding domain patterns from Ising model.

Figure 2 presents the simulated hysteresis loops with different average grain sizes. Each point in the hysteresis loops represents the average polarization at the end of 40000 MCSs at the given electric field. It shows that both the remanent polarization and the coercive field increase with the increase average grain size, also the increase of the polarization is much more pronounced than the increasing of coercive field. Accordingly from our model, the grain size has more influence on the remanent polarization rather than the coercive field. These features agree with the experimental cases in SBN[19] and PZT-20NZF[20] well. The microwave sintering of SBN[19] samples shows a remarkable increase in remanent polarization with the increasing grain size. The effect of grain size on the ME coefficient of PZT–20 NZF particulate composites has been investigated.[20] It was found that as the grain size increases, the overall area of the hysteresis loop is observed to enlarge, due primarily to an increase in the remanent polarization and the coercive field.

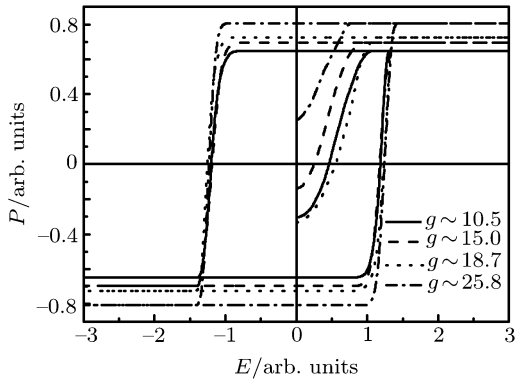


Fig.2. Hysteresis loops for different average sizes.

To illustrate the domain evolution process during the polarization reversal process, we plot domain patterns and the corresponding hysteresis loop in Fig.3 for sample with average grain size $g \sim 15$. The snapshots shown in Fig.3(b) are taken at different points marked in the hysteresis loop in Fig.3(a). As the applied external field reaches its maximum value, i.e., point (i) of Fig.3(a), we can see that the polarizations' directions of the domain in different grains are nearly in the same direction of the applied external field, no reversed domain appears in snapshot (i) of Fig.3(b). Normally the directions of the

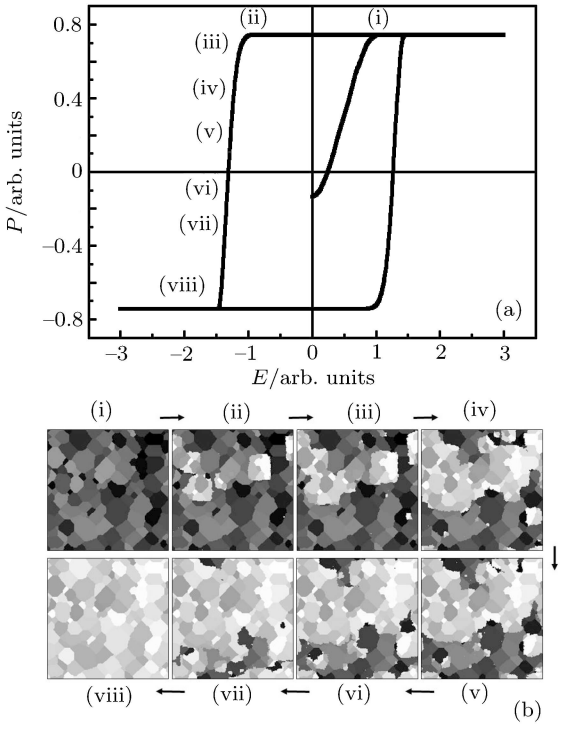


Fig.3. Domain patterns (b) at different stages of polarization reversal process (a).

grains do not completely parallel to the direction of the electric field, so the colour of domain in these grains changes from dark grey to light grey in the snapshots. As the electric field is reversed, see point (ii) of the hysteresis loop, the polarization begins to reverse in some grains, see the white portion in snapshot (ii) in Fig.3(b). It is shown that grain boundaries act as one of the sources for the nucleation of new domains during switching. After the polarization in these grains is reversed completely, polarization in their neighbouring grains is also reversed. This trend still keeps when the electric field increases in the negative direction, the polarization in more grains is reversed, also nucleation of reversed domain happens at different grains, see snapshots (iii) and (iv). The polarization reversion nearly is finished one grain by another, forming a region which covers several grains with polarization roughly along the direction of the electric field. We call this region as "extended domain", since the direction of the polarization is not exactly the same in the region because of the different orientations of the grains. The extended domain is more obvious when the electric field is near the coercive field, see snapshots (iv) and (v). As the electric field increases further, the extended domains merge, see snapshots (vi) and (vii). Finally the polarizations in all grains are almost reversed to the direction of the electric field, forming a single extended domain as shown in snapshot (viii). The light grey in snapshot (viii) means that the polarizations align in opposite direction of polarization in snapshot (i).

4. Conclusions

In this paper, we present how to use Potts–Ising model to simulate the switching properties of polycrystalline ferroelectrics. The grain texture, domain patterns and the polarization switching process are shown as example to demonstrate the validity of this scheme. Other properties of the coercive field such as remnant polarization, can be also obtained from this scheme. In addition, we found that the extended domain has been formed during the polarization reversal. This new scheme gives an insight into the relationship between the polycrystalline texture with different average grain size and the switching properties and is a valuable tool for exploring the physical properties of polycrystalline ferroelectrics.

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