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An inverse-pole-figure method for the analysis of domain switching in polycrystalline ferroelectrics/ferroelastics

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An inverse-pole-figure (IPF) method is proposed to analyze domain switching in polycrystalline ferroelectrics/ferroelastics subjected to electric/mechanical loading. The IPF method fixes the crystallite coordinates and makes electric/mechanical loading directions vary. Both the accuracy and efficiency of the IPF method are examined and proved superior to the conventional random statistic (RS) method. Furthermore, using the IPF method, it is very convenient to account for the depolarization field/stress caused by domain switching, thus predicting the observed incomplete switching in real ceramics.

Keywords: Inverse pole figure; Domain switching; Ferroelectrics; Ferroelastics; Polycrystalline

Ferroelectric materials have been widely used in sensors, actuators, transducers, etc., due to their peculiar electromechanical properties, ultrafast response and compact size. In practice, ferroelectric polycrystalline (or ceramics) are mostly preferred to single crystals due to their superior mechanical properties and lower cost. The constitutive response of ferroelectric polycrystalline under electric/mechanical loading requires more attention due to the complex domain switching effect involved. In the existing literature, a ferroelectric polycrystalline is usually made up of randomly oriented grains (or domains) and domain switching is analyzed using a random statistic (RS) method [1–3]. Generally, a large number of grains are required in the RS method to remove the errors by the pseudo-randomly generated grain orientations [2,3]. In this paper, we will show that a proposed inverse-pole-figure (IPF) method is quite efficient for analysis of polycrystalline ferroelectrics/ferroelastics. It can get the equivalent accuracy as the RS method using two-order less grain numbers. Furthermore, using the IPF method, it is very convenient to address the depolarization field/stress caused by domain switching.

The proposed IPF method is enlightened by the definition of the inverse-pole-figure in texture analysis [4]. For both tetragonal and rhombohedral ferroelectrics, the spontaneous strain $\boldsymbol{\varepsilon}^s$ is transversely isotropic around the polarization axis and can be expressed as

$$\boldsymbol{\varepsilon}^s = S_0\left(\mathbf{e_P}\,\mathbf{e_P} - \frac{1}{3}\mathbf{1}\right) \tag{1}$$

where $\mathbf{e_P}$ is the unit vector along the polar direction and $\mathbf{1}$ is the rank-2 identity tensor. S_0 is the single crystal deformation [5], $S_0 = S_{\text{lattice}} = c/a - 1$ for tetragonal ferroelectrics and $(8/9)S_0 = S_{\text{lattice}} = d_{[1\,1\,1]}/d_{[1\,1\,\bar{1}]} - 1$ for rhombohedral. Thus for a tetragonal (or rhombohedral) polycrystalline subjected to uniaxial electric/mechanical loading, as far as the polarization and strain are concerned, it is equivalent to fix the crystallite coordinates (x–y–z in Fig. 1) and make the field direction vary [6]. In the IPF method, each grain in a ferroelectric polycrystalline is made up of N ($N = 6$ for the tetragonal, $N = 8$ for the rhombohedral) types of domains. Because of symmetry, it is necessary to consider the case when the field direction falls on the shaded area of Figure 1. It should be noted that the IPF method proposed in this paper is based on the symmetric properties of spontaneous strain and polarization in tetragonal/rhombohedral ferroelectrics. It is used to study the domain switching process under applied field in polycrystalline ferroelectrics, while the uniform inverse-pole-figure in

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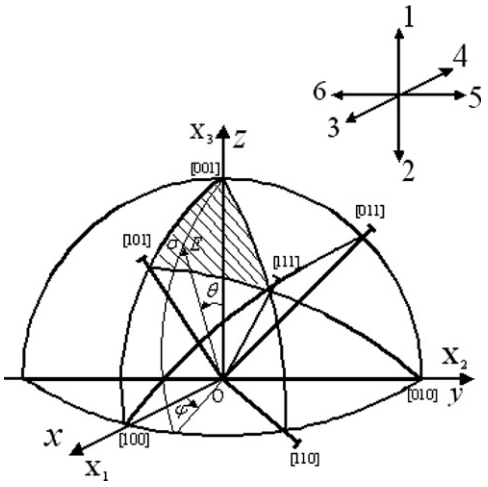

Figure 1. Illustration of the IPF method and six types of domains in tetragonal ferroelectrics.

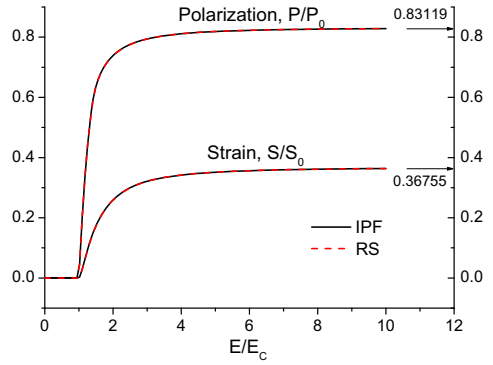

Figure 2. Simulated polarization and strain curves of tetragonal ferroelectric polycrystalline using IPF and RS methods with grain number $N = 90{,}000$.

Ref. [6] is only capable of solving the static, saturated domain orientation textures under infinite large electric field or stress.

Consider the case of an unpoled tetragonal ceramics during electric poling. Using an energy-based domain switching criterion similar to Hwang et al. [1] and switching option by Berlincourt and Krueger [7], when an electric field is applied at the shaded area of Figure 1, Domain 2 will switch to Domain 1 via 180° switching; Domains 4 and 6 will switch to Domain 1 via 90° switching first, then Domains 3 and 5 switch to Domain 1 via 90° switching. After getting the fractions of each type of domain switching, the polarization and longitudinal strain induced by domain switching can be explicitly obtained using integrations over the shaded spherical area in Figure 1, as expressed in Eq. (2):

$$P(E) = \frac{48}{4\pi}\int_0^{\pi/4} \mathrm{d}\varphi \int_0^{arc\cot(\cos\varphi)} \times P_0[2f_{2-1}z + f_{4-1}(z+x) + f_{6-1}(z+y) + f_{3-1}(z-x) + f_{5-1}(z-y)]\sin\theta \mathrm{d}\theta \tag{2a}$$

$$S(E) = \frac{48}{4\pi}\int_0^{\pi/4} \mathrm{d}\varphi \int_0^{arc\cot(\cos\varphi)} \times S_0[(f_{4-1}+f_{3-1})(z^2-x^2) + (f_{6-1}+f_{5-1})(z^2-y^2)]\sin\theta \mathrm{d}\theta \tag{2b}$$

where $x = \sin\theta\cos\varphi$, $y = \sin\theta\sin\varphi$, $z = \cos\theta (x, y, z \geqslant 0)$; P_0 is the magnitude of spontaneous polarization, f_{i-j} denotes the fractions of switching from Domain i to Domain j.

Figure 2 shows the simulated polarization and strain curves of tetragonal ferroelectric polycrystalline subjected to electric poling using the IPF method and RS method (where E_C is the coercive field). In the simulations, interactions between domains are neglected. The energy barriers for 180° and 90° domain switching are set to be $2P_0\ E_C$ and $\sqrt{2}P_0E_C$, respectively, to activate both types of switching at E_C. It can be seen that the simulation results by the two methods are completely overlapped, and both the polarization and strain approach the theoretical saturated values [6,8,9] with the increasing field.

To examine the accuracy and efficiency of the IPF method, the saturated polarization and strain of tetragonal and rhombohedral ceramics after poling are calculated and compared to that by the RS method, as shown in Table 1. It can be seen that, by averaging the errors for both types of ceramics, the accuracy of the IPF method using 1024 grains is almost equivalent to that of the RS method using 90,000 grains, which shows that the error of the IPF method caused by numerical integration is smaller than the RS method caused by pseudo-random number generation. Besides, the RS method always has a problem to realize the initial, unpoled state with zero polarization and strain [3]. Note that for the same grain number simulation, the time exhausted by the IPF method is about 80% of that by the RS method. All these indicate that the IPF method is obviously superior to the RS method in terms of accuracy and efficiency.

Using the IPF method, it is convenient to address the problems where the depolarization field caused by domain switching is taken into account; thus switching is a gradual process with the increasing field and is often incomplete. This is difficult, however, for the RS method where usually each grain is assumed to be a single domain [1–3]. Bearing in mind the charge screening effect in real ferroelectrics [10–12], the depolarization electric field by domain switching can be fully compensated by free charges. The depolarization stress field by 90° switching, however, cannot be compensated in a similar manner and it is acceptable to assume it proportional to the fraction of switched 90° domains [13]. If further assuming Domains 4 and 6 always switch to Domain 1 at same fractions (abbreviated as combined switching), the switching criterion for Domains 4 and 6 to Domain 1 can be written as follows:

$$EP_0(2\cos\theta + \sin\theta\cos\varphi + \sin\theta\sin\varphi) - kP_0E_C \cdot f = 2\sqrt{2}P_0E_C \tag{2}$$

where f is the fraction of completed 90° switching; k measures the level of depolarization field.

Figure 3 shows the simulated polarization and strain curves of $BaTiO_3$ ceramics during poling using different

Table 1. Saturated polarization and longitudinal strain in (a) tetragonal and (b) rhombohderal ferroelectric ceramics: values and relative errors (in parentheses) by the IPF method and the RS method using different grain numbers

Method	Grain number	Polarization (P/P_0)	Strain (S/S_0)
(a) Tetragonal ceramics			
IPF	1024	0.83126 (0.01%)	0.36767 (0.03%)
	10,000	0.83120 (0.001%)	0.36756 (0.003%)
	90,000	0.83118 (0.001%)	0.36756 (0.003%)
RS	1024	0.83376 (0.3%)	0.37126 (1.3%)
	10,000	0.83063 (0.05%)	0.36653 (0.3%)
	90,000	0.83117 (0.002%)	0.36753 (0.006%)
Accurate values (Ref.[6])		0.83119	0.36755
(b) Rhombohedral ceramics			
IPF	1024	0.86599 (0.005%)	0.42436 (0.014%)
	10,000	0.86602 (0.001%)	0.42441 (0.002%)
	90,000	0.86602 (0.001%)	0.42441 (0.002%)
RS	1024	0.86179 (0.5%)	0.41664 (1.9%)
	10,000	0.86707 (0.1%)	0.42610 (0.4%)
	90,000	0.86610 (0.01%)	0.42456 (0.08%)
Accurate values (Ref.[6])		0.86603	0.42442

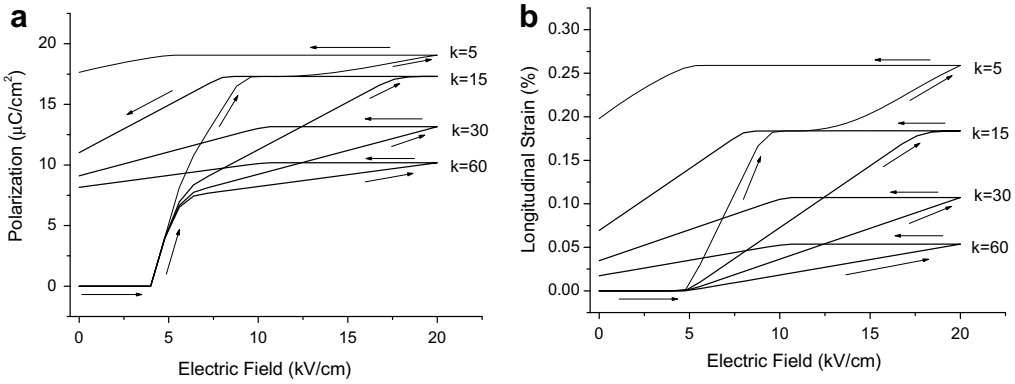

Figure 3. (a) Polarization and (b) strain curves of $BaTiO_3$ ceramics using different levels of depolarization field.

levels of depolarization field represented by varied k. In the simulations, the material parameters, $E_C = 4$ kV cm^{-1}, $P_0 = 26$ μC cm^{-2}, $S_0 = 0.01$, are used. It can be seen that when $k = 5$, the depolarization field is weak, thus combined 90° switching from Domains 4 and 6 to Domain 1 can be completed at about 9 kV cm^{-1}. Then no domain switching occurs until at about 12 kV cm^{-1}, when combined switching from Domains 3 and 5 to Domain 1 activates. The descending polarization/strain curves upon removing the electric field indicates that back switching occurs as a result of the depolarization stress field caused by 90° domain switching. When k is larger than 15, only the former type of combined 90° switching can occur under maximum field of 5 E_C. As the reported remnant polarization and strain in poled $BaTiO_3$ are about 8 μC cm^{-2} and 0.02% [7,14], respectively, it can be seen from Figure 3 that $k = 60$ is close to the real case.

Similarly, the IPF method can be used for ferroelastic ceramics under uniaxial mechanical loading. In ferroelastic case, only three types of domains exist in tetragonal crystals which can be denoted by 1, 3, 5 in Figure 1. Making the mechanical loading directions vary on the shaded area of Figure 1, domain switching in a ferroelastic polycrystalline can also be analyzed using a switching criterion similar to Eq. (2). Figure 4 shows the simulated stress–strain curves of an unpoled $BaTiO_3$

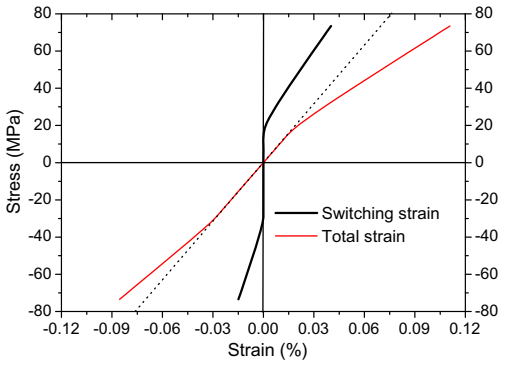

Figure 4. Simulated stress–strain curve of unpoled $BaTiO_3$ ceramics using IPF method.

ceramics under uniaxial tension and compression using the IPF method with the depolarization field level $k = 60$. The material shows linear behavior up to almost 40 MPa under compression and up to less than 20 MPa under tension, which fits the experimental observations very well [14].

From its definition, it can be deduced that the IPF can also be used to analyze domain switching in rhombohedral ceramics and morphotropic ceramics (where tetragonal and rhombohedral phases coexist [15]) subjected to coupled uniaxial electric/mechanical loading. In fact, the IPF method is valid for all the cases where domain textures can be fully characterized by a single inverse-pole-figure. Also, the IPF method is applicable to other ferroic polycrystalline materials such as ferromagnetics, shape memory alloy [16], etc.

In summary, we proposed an IPF method to analyze domain switching in ferroelectric/ferroelastic polycrystalline. Both the accuracy and efficiency of the IPF method are proved to be superior to the conventional random statistic (RS) method. Furthermore, using the IPF method, the depolarization field effect caused by domain switching is convenient to be taken into account while it is difficult to use the RS method.

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[1] S.C. Hwang, C.S. Lynch, R.M. McMeeking, Acta. Metall. Mater. 43 (1995) 2073.
[2] F.X. Li, D.N. Fang, Mech. Mater. 36 (2004) 959.
[3] F.X. Li, D.N. Fang, A.K. Soh, Scripta Mater. 54 (2006) 1241.
[4] H.J. Bunge, Texture Analysis in Materials Science, Butterworth, Berlin, 1982.
[5] B. Jaffe, W.R. Cook, H. Jaffe, Piezoelectric Ceramics, Academic Press, London/New York, 1971.
[6] F.X. Li, R.K.N.D. Rajapakse, J. Appl. Phys. 101 (2007) 054110.
[7] D. Berlincourt, H.H.A. Krueger, J. Appl. Phys. 30 (1959) 1804.
[8] H.G. Baerwald, Phys. Rev. 105 (1957) 480.
[9] N. Uchida, T. Ikeda, Jpn. J. Appl. Phys. 6 (1967) 1079.
[10] M.E. Lines, A.M. Glass, Principles and Applications of Ferroelectrics and Related Materials, Clarendon Press, Oxford, 1977.
[11] D.C. Lupascu, Fatigue in Ferroelectric Ceramics and Related Issues, Springer-Verlag, Berlin/New York, 2004.
[12] D.A. Hall, A. Steuwer, B. Cherdhirunkorn, P.J. Withers, T. Mori, J. Mech. Phys. Solids 53 (2005) 249.
[13] S. Wan, K.J. Bowman, J. Mater. Res. 16 (2001) 2306.
[14] E.C. Subbarao, M.C. McQuarrie, W.R. Buessem, J. Appl. Phys. 28 (1957) 1194.
[15] B. Noheda, J.A. Gonzalo, L.E. Cross, R. Guo, S.E. Park, D.E. Cox, G. Shirane, Phys. Rev. B 61 (2000) 8687.
[16] K. Bhattacharya, R.V. Kohn, Acta Mater. 44 (1996) 529.