A new insight into structural complexity in ferroelectric ceramics

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Abstract: The structure of the ferroelectrics has been widely studied in order to pursuing the origin of high electromechanical responses. However, some experiments on structure of ferroelectrics have yielded different results. Here, we report that the controversial phase structure is due to the adaptive diffraction of nanodomains which hides the natural crystal structure, and the electric-field-induced phase transition is that the natural crystal structure reappears due to the coalescent nanodomains or ordering nanodomains by applying a high electric field. The temperature dependence of dielectric constant with different measurement frequencies and X-ray diffraction (XRD) patterns of unpoled, poled, and annealing after poled ceramics in $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3\text{--BaTiO}_3$ (BNT–BT) ceramics authenticate the statement. These results provide a new insight into the origin of structural complexity in ferroelectric ceramics, which is related to the key role of nanodomains.

Keywords: crystal structure; adaptive diffraction; ferroelectricity; phase transitions

1 Introduction

Perovskite ferroelectric (FE) materials with excellent electromechanical and dielectric properties are widely used as sensors, actuators, generators, and ultrasound transducers [1–3]. Knowledge of the natural crystal structure distortions in ferroelectric is a fundamental issue to understanding the physical and structural origin of enormous electromechanical and high piezoelectric responses under a high electric field that has remained elusive for decades [4–6]. Although most of the studies demonstrate that the ferroelectric solid solutions of $\text{Pb}(\text{Zr}_x\text{Ti}_{1-x})\text{O}_3$ (PZT), $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})_x\text{Ti}_{1-x}\text{O}_3$ (PMNT), $\text{Pb}(\text{Zn}_{1/3}\text{Nb}_{2/3})_x\text{Ti}_{1-x}\text{O}_3$ (PZNT), $(1-x)\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3\text{--}x\text{BaTiO}_3$ (BNT–BT), and $(\text{K},\text{Na})\text{NbO}_3$ (KNN) in the vicinity of morphotropic phase boundary (MPB)

owning to structure instability yield high piezoelectricity, the origin of the MPB and exact structure/symmetry at MPB region is still under controversy [7–16].

Over many years, it came to be generally accepted that the so-called MPB in PZT and BNT–BT is a region of coexistence between the tetragonal (*T*) and rhombohedral (*R*) phases. Shortly afterwards, a new low-symmetry bridging monoclinic phase at the MPB was reported based on X-ray diffraction experiment [17,18]. Interestingly, subsequent X-ray investigations revealed nothing but a cubic symmetry for compositions near the MPB in BNT–BT virgin state [20,21]. And further transmission electron microscopy (TEM) investigations have not found a domain structure [22]. Rao *et al.* [23] have pointed out that local heterogeneity has apparent influence on average global structure in $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ ceramics. Additionally, Bokov *et al.* [24] suggested that a remarkable

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peculiarity of PZT and relaxor-based solid solution is that the crystal symmetry may depend on the length scale under consideration.

It is noted that the existence of nanodomains is a common feature of the MPB region [18,19,25–31]. It has been reported that the nanodomains, whose size is much smaller than the coherence length of diffraction radiations, scatter waves from individual nanodomains coherently superimposed in diffraction profiles, and cause anisotropic broadening, asymmetries, intensity distribution variation between the peaks, and even new peaks appearance in diffraction patterns [31,32]. The adaptive ferroelectric states indicated that the monoclinic phase is just a rhombohedral and/or tetragonal nanodomains-averaged structure, which is not a real phase in the usual thermodynamic sense [26,33]. Therefore, conventional X-ray or neutron powder diffraction and corresponding Rietveld refinement can easily lead to erroneous phase structure if we do not take into account the effect of nanodomains on diffraction peaks.

Here, we presented a model with adaptive diffraction of nanodomains, where scattered waves from poly nanodomains coherently superimpose to form new Bragg reflection peaks, which may rationalize the structural complexity in ferroelectric polycrystalline. Our experimental data further demonstrated that the X-ray coherent diffraction of nanodomains hides the natural structure of nanodomain materials, and nanodomain rearrangement driven by electrical field would recur to the natural crystal structure, which was generally inappropriately accepted as electric-field-induced irreversible phase transition.

Our results naturally explain the controversial phase structure of BNT–BT systems in the MPB region and will provide a new insight into the interplay between giant dielectric, strain, ultrahigh piezoelectric responses, and microstructure in ferroelectrics.

2 Experimental procedure

The system we have studied is $(1-x)(\text{Bi}_{0.5}\text{Na}_{0.5})\text{TiO}_3-x\text{BaTiO}_3$ (BNT– x BT, $x=0.06$ and 0.11). Ceramic samples were prepared through a solid oxide route using reagent-grade elemental oxides and carbonates of BaCO_3 , Bi_2O_3 , TiO_2 , and Na_2CO_3 powders. The starting powders were mixed for more than 24 h before calcined at 900°C for 2 h. The calcined powders were

mixed with polyvinyl acetate and then were pressed in 1.2 cm diameter disk for sintering. The sintering conditions were 1150°C for 2 h. The X-ray diffraction patterns were recorded on unpoled and poled sintered ceramics by X-ray diffraction (XRD, Bruker D8-Advance) operated with $\text{Cu K}\alpha$ radiation. The permittivity was measured with an impedance analyzer (Agilent 4294A) interfaced with a computer to control temperature.

3 Results and discussion

Note that the X-ray diffraction experimental data can only give the structure factor (F_{HKL}) value, which is described as follow:

$$\begin{aligned}
 |F_{\text{HKL}}| &= \left\{ \left[\sum_{j=1}^n f_j \cos 2\pi(Hx_j + Ky_j + Lz_j) \right]^2 + \left[\sum_{j=1}^n f_j \sin 2\pi(Hx_j + Ky_j + Lz_j) \right]^2 \right\}^{1/2} \\
 &= \left[\left(\sum_{j=1}^n M_j \right)^2 + \left(\sum_{j=1}^n N_j \right)^2 \right]^{1/2} \\
 &= \{ [M(\text{A}) + M(\text{O}) + M(\text{Ti})]^2 + [N(\text{A}) \\
 &\quad + N(\text{O}) + N(\text{Ti})]^2 \}^{1/2} \quad (1)
 \end{aligned}$$

where f_j is the atomic scattering factor, A, O, and Ti are A site, oxygen, and titanium ions in ABO_3 -type perovskite ferroelectrics, respectively. In order to simplify the formula, we only consider the Ti^{4+} ion displacement in the TiO_6 -octahedron. Since the Ti^{4+} ion displacement is quite small, as shown in Fig. 1, thus the $M(\text{Ti})$ value is much larger than that of $N(\text{Ti})$. Simultaneously, $N(\text{Ti})$ value in Fig. 1(b) and $N(\text{Ti})$ value in Fig. 1(c) are equivalent, but their signs are opposite with the positive and negative signs. Thus, the structural models in Fig. 1(b) and Fig. 1(c) are very much alike in X-ray scale, and are indistinguishable from ion thermal vibration. If one considers half Ti^{4+} ions move up and the other half move down, there is a strong resemblance in structural model among Fig. 1(b), Fig. 1(c), and Fig. 1(d). These results strongly suggest that the conventional X-ray diffraction would be unable to detect the crystal structure of nano-sized domain ferroelectric materials.

Fig. 1 Four modes of Ti^{4+} ion displacement in the TiO_6 -octahedron.

Given that the existence of nanodomains in MPB region and the adaptive diffraction of nanodomains are inherent features in ferroelectrics, it is perhaps not surprising to propose a model which can indicate the

effect of nanodomains on average crystal structure in XRD patterns in ferroelectrics. Figure 2 shows the model for the effect of nanodomain structure on average crystal structure of polycrystalline ferroelectric ceramics. For diffraction analysis of ferroelectric materials, the diffraction conditions are required for a resolution that the diffraction spots can be discerned from adjacent domains. The resolution is determined by the size of domains, the value of the tetragonal (or rhombohedral) c/a ratio, and the interplanar distance of reflection spot. For ceramic materials with large size domains, domain sizes are larger than the coherence length of diffraction radiation, thus the conventional reflection peaks from individual macrodomain are the same as that of grains/bulk, and the local internal structure of macrodomains is the same as the average crystal structure of bulk ceramics (Fig. 2(a)). When the domain sizes are down to nanoscale, the diffraction

Fig. 2 Effects of local domain microstructure on the average structure in ferroelectrics. (a) Macrodomain materials with local domain structure and diffraction profiles of average structure. (b) Nanodomain materials with cubic diffraction peaks of average structure. (c) Materials with coexistence of macrodomains and nanodomains and the overlapping diffraction profiles of average structure. (d) Under applying an electrical field of nanodomain materials with the diffraction peaks of average structure.

spots from adjacent domains can not be distinguished by the XRD patterns, the conventional diffraction peaks from coarse domains would disappear while new adaptive Bragg reflection peaks of nanodomains appear (Fig. 2(b)) [17]. The resolution of X-ray diffraction can not afford enough capacity to detect the nanodomain symmetry. Thus, a cubic phase in ferroelectrics is observed for nanodomain materials regardless of the details of the microscopic symmetry of the nanodomains. This issue is consistent with the generally observed cubic symmetry in relaxor ferroelectrics with polar nanoregions (PNRs). When nanodomains coexist with large size domains, the superimposed peaks of the adaptive peak and the conventional Bragg peaks (Fig. 2(c)) can easily be interpreted as evidence of a new low symmetry phase or two-phase state.

On the other hand, when an electric field is applied on the nanodomain materials, these nanodomains would align along the direction of the external field. Thus, the nanodomains directly transform to thick lamellar domains (coarse domains) through the irreversible coalescence of individual nanodomains, or ordering which would cause an enhancement of the coherence length between individual nanodomains. When the coalescent domains or long-range ordering of nanodomains persist after external field removed, and their sizes are larger than the coherence length of diffraction radiation, the adaptive Bragg reflection peaks of nanodomains disappear, and the natural average symmetry of bulk ceramics appears which is accordant with the internal symmetry of nanodomains. Recently, Refs. [34–36] demonstrated that the observed electric field induced phase transformation behavior in BNT–BT is a consequence of the changes in the coherence lengths of the PNRs. Therefore, the widely accepted theory of field-induced phase transition may be a transition from adaptive diffraction of the nanodomains to conventional diffraction of the coarse domains. Furthermore, the adaptive phases consisting of mixed nanodomains are significantly sensitive to electric field, which is easily suggested as electric-field-induced phase transformation. However, the real ferroelectric phases with coarse domains are determined by atomic bonds, composition, and temperature, which would be weakly dependent on electric field [25], which will be proved in the following experimental result section.

Here, we define the model as a name of the nanodomains obscuring the natural crystal structure (NONCS) and the nanodomain ordering recurring the

natural crystal structure (NORNCS). The model with regard to the NONCS/NORNCS provides the appropriate background for understanding the origin of the structural complexity in diffraction patterns of PZT, PMNT, PZNT, and BNT–BT after different electric field and thermal treatments.

It is well known that the polycrystalline ferroelectric ceramics should be compositionally ordered and disordered with multi-scale inhomogeneities of domain structure from nanodomains to hierarchical structures and mosaic domain microstructure [37]. The relative fraction of the coexistence domain structure is highly susceptible to thermal, mechanical, and electrical stimuli. On the other hand, the coherence length of low-resolution and high-resolution X-ray and neutron diffraction is different from instruments, which would cause the multi-scale adaptive diffraction from the nanodomains to submicrodomains, even microdomains. The diffraction intensity ratio from the adaptive diffraction of small-scale domains and conventional diffraction of large-scale domains is diversely dependent on the relative fraction, size, and configuration of multi-scale domains, which should be the origin of the structural complexity of ferroelectrics. For example, on a length scale of X-ray diffraction, the average symmetry of the MPB region is generally detected as cubic/pseudo cubic phase when the microstructure is occupied dominantly by the nanodomains.

The domain topography and structure were general observed by transmission electron microscopy (TEM). However, the necessity to use very thin samples in this case may alter the microstructure of the domains and therefore the results may not be representative of the bulk. Recent studies on BNT–BT system established a clear relation between domain morphology and dielectric frequency dispersion in the frequency range of 1–100 kHz: nanodomains correspond to strong frequency dispersion while large domains show weaker frequency dependence [25]. Therefore, the domain evolution can be characterized by the dielectric frequency dispersion.

The electric field behavior of the nanodomains is illustrated using the $(1-x)(\text{Bi}_{0.5}\text{Na}_{0.5})\text{TiO}_3-x\text{BaTiO}_3$ (BNT– x BT, $x=0.06$ and 0.11) ceramics as an example, whose microstructure and crystal structure are widely studied and are still under controversy with several theories like the coexistence of R and T phases, the cubic phase, and the intermediate bridging monoclinic phase [11,12,20–22].

Figure 3 depicts the temperature dependence of dielectric permittivity (ϵ_r) under the frequencies of 1, 10, and 100 kHz, and the XRD patterns for both unpoled and poled BNT- x BT ceramics. It is clear that the unpoled specimen of BNT-0.06BT is nanodomain materials with strong dielectric frequency dispersion at room temperature, and irreversibly transforms into a coarse-domain or ordering domain ceramics with weak dielectric frequency dependence by applying an enough strong external electric field (50 kV/cm). A correlation between nanodomain converge to coarse domains versus crystal structure variation is represented in Fig. 3(b). The nearly cubic phase of virgin state polycrystalline ceramics was associated with average structure of the adaptive R and T nanodomains due to NORCS. During poling the domain sizes or the correlation length of the nanodomains increased, and real R and T phases appeared due to NORNCS. The results clearly indicate that the adaptive nanodomain materials are greatly sensitive to electrical field. Since

the electric induced coalescent domains or ordering nanodomains are metastable, the poled ceramics turn into the nanodomain state when the temperature increases to $\sim 140^\circ\text{C}$, and the observed average structure also returns to cubic phase (Fig. 3(b)). The metastable state implied that the electric induced transition phase is not equilibrium phase in thermodynamics sense. The difference of XRD profile between the unpoled and the annealed sample as shown in Fig. 3(b) may arise from the stress relief or stress accommodation of ordered domain retuning to virgin state during annealing treatment.

Comparing the temperature dependence of dielectric constant of poled and unpoled BNT-0.11BT samples, it clearly suggests that the ceramics are coarse-domain material ceramics with weak dielectric frequency dependence. The corresponding average crystal structure of bulks and local macrodomain structure were accordant and no obvious phase transition was observed to occur. Clearly, the conventional

Fig. 3 Temperature dependence of dielectric constant with 1, 10, and 100 kHz measurement frequencies and XRD patterns of unpoled, poled, and annealing after poled ceramics. (a) Temperature dependence of dielectric constant of unpoled and poled BNT-0.06BT ceramics. (b) XRD patterns of unpoled, poled, and annealing (at 140°C) after poled BNT-0.06BT ceramics. (c) Temperature dependence of dielectric constant of unpoled and poled BNT-0.11BT ceramics. (d) XRD patterns of unpoled and poled BNT-0.11BT ceramics.

macrodomain materials with real ferroelectric phase are fairly stable under external field.

The above models and results clearly demonstrate that the microstructure plays a crucial role in the structure analyses of ferroelectrics. The models and results are also adaptive to other domain materials.

4 Conclusions

The observed cubic phase in ferroelectrics is due to the adaptive phase of the nanodomains. The generally accepted electric-field-induced phase transition is actually the reappearance of real crystal structure due to the adaptive phase (nanodomain) transforms to conventional phase under high electric field (coalescent domains or long-range ordering of nanodomains). The coexistence of complex domain structure in polycrystalline ferroelectric ceramics gives rise to the structural complexity in ferroelectrics. We believe that our results will stimulate further work on researching the structure–property correlation in ferroelectrics.

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