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Dielectric properties and phase transition of PMN0.32PT single crystal under dc electric field

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Abstract

The temperature dependence of dielectric properties for a PMN0.32PT single crystal under different dc electric fields (E) has been investigated. The dielectric responses and phase transition behaviors of this material are provided. With an increasing electric field, both phase transition temperatures of rhombohedral to tetragonal phase and tetragonal to cubic phase increased. When $E = 1.5$–4.0 kV/cm, a new phase is induced by an electric field. The phase transition and the induced new phase were discussed.

Keywords: PMNPT single crystal; Dielectric properties; Phase transition; dc electric field

1. Introduction

Single crystal relaxor ferroelectrics $(1-x)Pb(Mg_{1/3}Nb_{2/3})O_3$–$xPbTiO_3$ (PMN–xPT), especially composition near morphotropic phase boundary (MPB), where $0.28 < x < 0.36$, have been reported to exhibit much large piezoelectric constants, electromechanical coupling factors and electrostrictive coefficient [1,2]. The macroscopic properties of a PMN–PT single crystal are known to depend closely on the external applied electric filed, e.g. ferroelectric, electrostrictive. Park et al. [1], Liu et al. [3], and Fu and Cohen [4], attributed the ultrahigh strain under electric field in these single crystals to the phase transition from rhombohedral to tetragonal or the polarization rotation induced by the electric field. Therefore it is necessary to study electric field-induced phase transition in PMN–PT single crystals.

Sommer et al. [5] studied the electric field and temperature effects on the dielectric constant and depolarization current and proposed a phase diagram delimiting the dynamic disorder, glassy, metastable ferroelectric and induced polarization states. An electric field-induced first-order phase transition from the disorder cubic phase to a macroscopically polar 3m phase was disclosed in high quality single crystal. Tu et al. [6] found the extra peak at 370 K in dielectric permittivity for PMN–24%PT single crystal in the zero-field-heated after field cooling (FC-ZFH) under a dc bias field. They thought that the extra peak is attributed to the establishment of the long-range percolating ferroelectric cluster during the FC process, similarly to the extra dielectric peak at ~212 K in PMN single crystal. Because of separating rhombohedral (or pseudocubic) and tetragonal phases for

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compositions lying near MPB in PMN–PT system, the dielectric behavior and phase transition of such single crystals are complex under dc field.

Since Noheda et al. [7], discovered monoclinic phase at MPB in PZT system in 1999, much attention has been given to study the new phase in single crystals of PZN–PT and PMN–PT system near the MPB. Upon application of an electric field along $[0\,0\,1]_{cubic}$, evidence of polarization rotation via a monoclinic phase has been shown for PZN–8%PT [8]. Recently, a monoclinic phase has been discovered in 0.65PMN–0.35PT single crystal by means of high-resolution synchrotron X-ray diffraction. It appears at room temperature in a single crystal previously poled under an electric field of 43 kV/cm applied along the pseudocubic [0 0 1] direction, in the region of the phase diagram around the MPB between the rhombohedral and tetragonal phase [9]. After discovery of monoclinic phase, it will be more interesting and complex to study the dielectric properties and phase transition under a dc field.

In this paper, the temperature dependence of dielectric properties for $0.68Pb(Mg_{1/3}Nb_{2/3})O_3$–$0.32PbTiO_3$ (PMN0.32PT) single crystal under different dc electric field (E) has been investigated. The dielectric responses and phase transition of this material are provided and discussed.

2. Experiment

The single crystal of the composition PMN0.32PT was grown using an accelerated crucible rotation technique and Bridgman method [10]. The single crystal was oriented along pseudocubic [0 0 1] directions by XRD. The sample of size of $5 \times 5 \times 0.8$ mm was used for dielectric measurements. The applied electric field was along the [0 0 1] direction. Dielectric measurements were conducted on an automated system, where a computer controlled a Delt temperature box, a HP4284A LCR meter and a high voltage generator. Dielectric properties were measured at 0.1, 1, 10, 100 kHz, in the temperature range from room temperature to 200 °C with a heating rate of 3 °C/min. Due to the sample history has a strong influence on the dielectric response, the sample was annealed at 300 °C for 2 h before dielectric measurement. For the field-heated (FH), the dielectric properties were measured under a dc field while heating.

3. Results and discussion

Fig. 1(a)–(d) show the temperature dependence of the dielectric constant and dissipation factor under 0, 1.5, 2 and 4 kV/cm by dielectric measurement of FH, respectively. The temperatures of T_m, corresponding to ferroelectric to paraelectric phase transition, are at 138.4 , 142, 143.7 and 148 °C at 1 kHz under 0, 1.5, 2 and 4 kV/cm, respectively. The temperatures of the rhombohedral to tetragonal phase transition are at 80.5, 90.6, 91.2 and 100.3 °C, respectively. When $E = 1.5$ and 2 kV/cm, the abnormal dielectric peaks appear in rhombohedral phase region. Corresponding to the curve of tg δ vs. T, there are sharp peaks at around 70 °C. When $E = 4$ kV/cm, the abnormal dielectric peaks are not evident and degenerate into a turning point. However corresponding to the curve of tg δ vs. T, there is a sharp peak at about 90 °C. Fig. 2 shows the temperature dependence of dielectric constant under different dc field at 1 kHz by dielectric measurement of FH. It is indicated that the two kinds of phase transition temperatures and the temperature of abnormal dielectric peaks increased and dielectric constants decreased with increasing dc field.

We have reported the phase transition process in PMN0.32PT single crystal without dc field in a separated paper [10]. In Fig. 1(a), a paraelectric phase is transformed into a weak relaxor at $T = T_m$. The ferroelectric microdomains exist between T_m and T_c. When $T = T_c$, a first-order transformation from weak relaxor ferroelectrics to normal ferroelectrics happens, corresponding to a polar micro–macrodomain switching. In Fig. 1(b)–(d), we find that the phase transition behavior from ferroelectric to paraelectric phase under dc field is similar to that without dc field. When $T > T_m$, the frequency dispersion still exists. This dielectric behavior is different from normal ferroelectrics. With an increasing dc field, the temperatures of phase transition increased, at the same

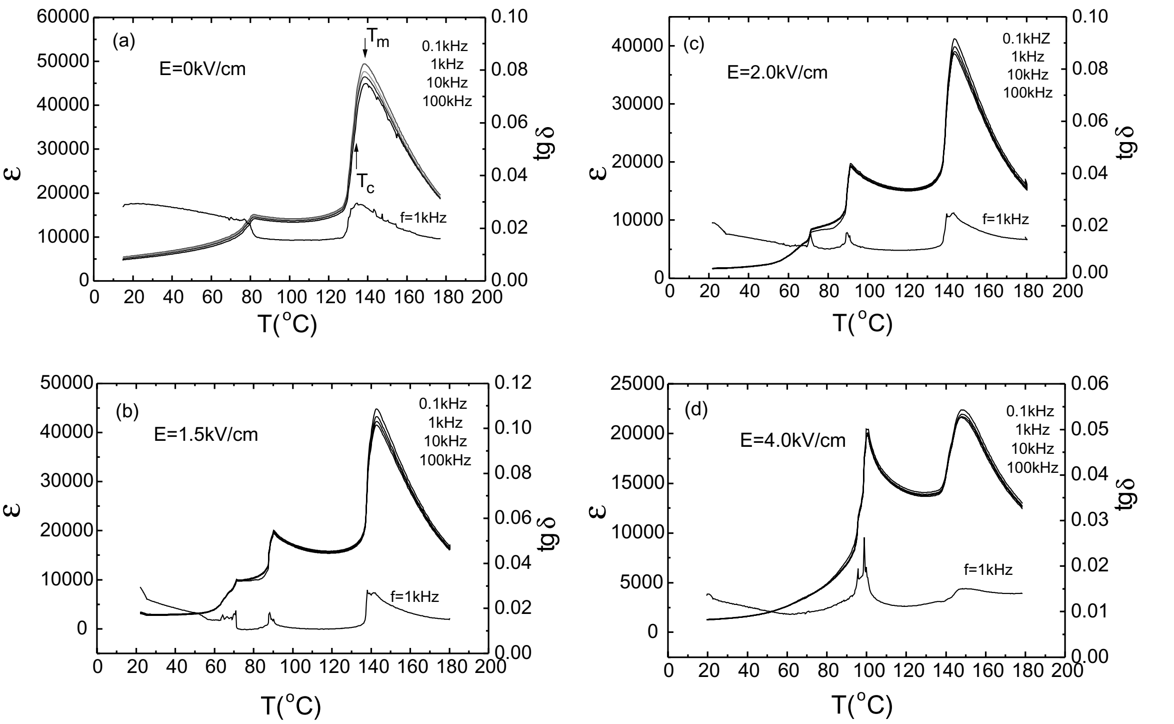

Fig. 1. Temperature dependence of dielectric constant and dissipation factor under (a) 0 kV/cm, (b) 1.5 kV/cm, (c) 2 kV/cm and (d) 4 kV/cm (FH).

time, ε reduced largely. The temperature stable range of tetragonal phase decreased. When dc field is zero, the temperature range of tetragonal phase is about 58 °C. When dc field is 8 kV/cm, the range is 32 °C. Both effects are associated with the

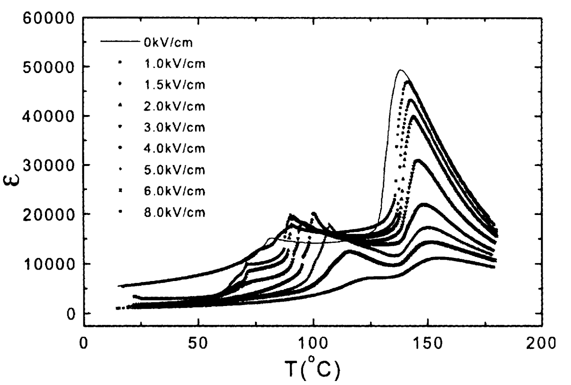

Fig. 2. Temperature dependence of dielectric constant under different dc field at 1 kHz (FH).

alignment of polar domains with the applied dc field, which implies (i) the need for more thermal energy to disrupt the alignment (thus a high T_c) and (ii) lower lattice polarizability (thus lower ε [11].

According to the studies of Noheda et al. [8] upon application of an electric field along $[0\,0\,1]_{cubic}$, evidence of polarization rotation via a monoclinic phase has been shown for PZN–8%PT. Ye et al. [9] reported the results of a high-resolution synchrotron X-ray diffraction study of 0.65PMN–0.35PT single crystals, showed unambiguous evidence of a monoclinic phase, which was stable between the rhombohedral and tetragonal phases near the MPB. The phase diagram shows in Fig. 3. So we suppose that the abnormal dielectric peaks may correspond to the phase transition from rhombohedral to monoclinic phases under dc electric field. When dc field is low or even to zero, there are not abnormal dielectric peaks. It indicates that dc field is too small to induced the phase

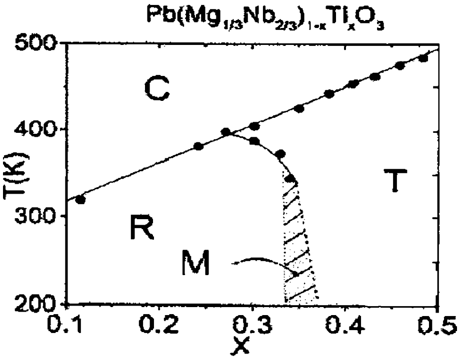

Fig. 3. Phase diagram of PMN–PT solid solution system [9].

transition from rhombohedral to monoclinic during heating process. When dc field is 1.5–4 kV/cm, the induced phase transitions take place in PMN0.32PT single crystal with heating. However, when dc field is higher than 4 kV/cm, the abnormal dielectric peaks disappear. It shows that the dc field is high enough to result in the polarization rotation quickly and the induced phase transition from rhombohedral to monoclinic phases is not exhibited at ε–T curve during heating process. That is to say, application of high electric field and heating make the polarization rotation quickly and the temperature stable regions of monoclinic phase disappear. The electric field–temperature (E–T) phase diagram is shown in Fig. 4.

In PMN–PT or PZN–PT system, spontaneous polarization is in [1 1 1] direction. If a dc field is applied along [0 0 1], the polarization rotation takes place from rhombohedral to tetragonal phases via a monoclinic phase. Ye et al. [9] found a monoclinic phase in a 0.65PMN–0.35PT single crystal poled under an electric field of 43 kV/cm applied along the pseudocubic [0 0 1] direction previously. However, in our experiments, the monoclinic phase is induced under a 1.5–4 kV/cm dc field during heating process. It shows that an external electric field lowers if the sample is heating at the same time, that is to say, the monoclinic phase is induced easily under dc field while heating.

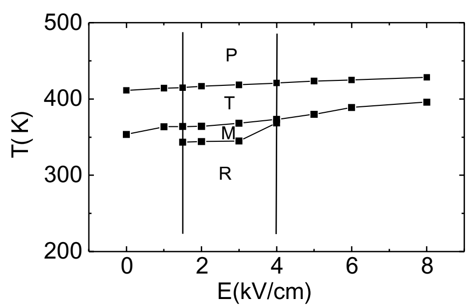

Fig. 4. The electric field–temperature (E–T) phase diagram of PMN0.32PT single crystal.

4. Conclusions

The temperature dependence of dielectric properties for a PMN0.32PT single crystal under different dc electric fields (E) have been investigated. When $E = 1.5$–4.0 kV/cm, a monoclinic phase is induced by an electric field during heating process. The phase transition can be shifted to high temperatures by an increasing electric field.

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