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Direct observation of domain wall motion and novel dielectric loss in 0.23Pb(In_{1/2}Nb_{1/2})O₃-0.42Pb(Mg_{1/3}Nb_{2/3})O₃-0.35PbTiO₃ crystals

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Direct observation of domain wall motion and novel dielectric loss in $0.23\text{Pb}(\text{In}_{1/2}\text{Nb}_{1/2})\text{O}_3\text{-}0.42\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-}0.35\text{PbTiO}_3$ crystals

Abstract

Domain wall motion was directly observed at temperatures near to the monoclinic-to-tetragonal phase transition temperature ($T_{\text{M-T}}$) in [001]-oriented $0.23\text{Pb}(\text{In}_{1/2}\text{Nb}_{1/2})\text{O}_3\text{-}0.42\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-}0.35\text{PbTiO}_3$ ($0.23\text{PIN}\text{-}0.42\text{PMN}\text{-}0.35\text{PT}$) single crystals using a polarizing light microscope, and evident 90° and 180° domain switches were observed near the Curie temperature (T_{C}). Two dielectric loss anomalies were observed at temperatures near $T_{\text{M-T}}$ in the [001]-oriented PIN-PMN-PT single crystals, while an additional dielectric loss peak was found at temperatures a few degrees below T_{C} , which was associated with domain wall motion. Based on the domain structure observations, a domain switching mechanism was proposed to explain the novel dielectric loss peak at several degrees below T_{C} . 2013 The Royal Society of Chemistry.

Keywords

novel, dielectric, $0.23\text{Pb}(\text{In}_{1/2}\text{Nb}_{1/2})\text{O}_3\text{-}0.42\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-}0.35\text{PbTiO}_3$, crystals, motion, loss, wall, domain, observation, direct, 0.23pb , $\text{in}_{1/2}$, $2\text{nb}_{1/2}$, 0.35pb , $\text{mg}_{1/3}$, $\text{nb}_{2/3}$

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Domain wall motion was directly observed at temperatures near to the monoclinic-to-tetragonal phase transition temperature (T_{M-T}) in [001]-oriented 0.23Pb(In_{1/2}Nb_{1/2})O₃–0.42Pb(Mg_{1/3}Nb_{2/3})O₃–0.35PbTiO₃ (0.23PIN–0.42PMN–0.35PT) single crystals using a polarizing light microscope, and evident 90° and 180° domain switches were observed near the Curie temperature (T_C). Two dielectric loss anomalies were observed at temperatures near T_{M-T} in the [001]-oriented PIN–PMN–PT single crystals, while an additional dielectric loss peak was found at temperatures a few degrees below T_C , which was associated with domain wall motion. Based on the domain structure observations, a domain switching mechanism was proposed to explain the novel dielectric loss peak at several degrees below T_C .

Introduction

Domain wall motion has a dominant position in the macroscopic response of ferroelectric and ferroelastic materials, for example, in the complex nonlinear response and significant hysteresis. In particular, it has been reported that the dielectric properties are closely related to domain wall motion in ferroelectric materials,^{1–3} where a dielectric loss peak has been observed at temperatures a few degrees below the Curie temperature (T_C), which is thought to be related to the movement of the domain walls. However, there is no direct observation of domain wall motion associated with the dielectric loss peak. It is one purpose of this research to study the domain wall evolution at various temperatures in order to understand the contribution of domain wall motion to the macroscopic dielectric properties.

Recently, Pb(In_{1/2}Nb_{1/2})O₃–Pb(Mg_{1/3}Nb_{2/3})O₃–PbTiO₃ (PIN–PMN–PT) ternary crystals with a higher T_C than that of Pb(Mg_{1/3}Nb_{2/3})O₃–PbTiO₃ binary crystals, and comparable high piezoelectric and dielectric properties near the morphotropic

phase boundary (MPB) compositions, have attracted extensive investigations.^{4–6} The high piezoelectric coefficient d_{15} in the single domain tetragonal PIN–PMN–PT crystals and the high extensional piezoelectric coefficients d_{33}/d_{32} in the multi-domain PIN–PMN–PT crystals were thought to be related to the domain state and polarization rotation. From the viewpoint of application, it is desirable to investigate the evolution of the domain structure at various temperatures in the PIN–PMN–PT crystals. In this work, the temperature induced phase transitions were investigated in the [001]-oriented 0.23Pb(In_{1/2}Nb_{1/2})O₃–0.42Pb(Mg_{1/3}Nb_{2/3})O₃–0.35PbTiO₃ (0.23PIN–0.42PMN–0.35PT) crystals based on *in-situ* domain structure observations and dielectric/ferroelectric property measurements. A domain switching model was proposed to explain the electrical properties.

Experimental

PIN–PMN–PT crystals were grown using a modified Bridgman method. The studied composition was 0.23PIN–0.42PMN–0.35PT, which is close to the monoclinic to tetragonal MPB region.⁷ The samples with sizes of 5 × 5 mm² were polished to 70 μm. The domain structure was observed using a polarizing light microscope (PLM) (Olympus BX51) with a LINKAM heating–cooling stage. Prior to the dielectric measurements, the samples were sputtered with transparent electrodes and poled at an electric field of 8 kV cm^{−1} at room temperature. The dielectric permittivity and dielectric loss were measured under a zero bias field as a function of temperature in the range of 20 °C to 250 °C at a heating rate of 3 °C min^{−1}, by a multi-frequency LCR meter (HP4284A), which was connected to a computer controlled furnace. The polarization-field hysteresis loops were measured using a TF analyzer 2000 (aixACCT).

Results and discussion

The domain structure vs. temperature was investigated in [001]-poled 0.23PIN–0.42PMN–0.35PT crystals by *in-situ* observations. The crystals presented two types of domains at 30 °C, as shown in Fig. 1(a). One domain had domain walls parallel to the [010]

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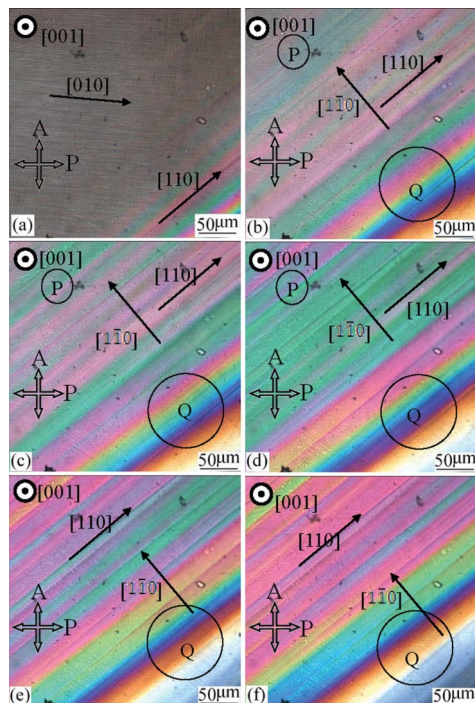


Fig. 1 The domain structures observed in the [001]-oriented 0.23PIN–0.42PMN–0.35PT crystals at temperatures of (a) 30 °C, (b) 40 °C, (c) 50 °C, (d) 60 °C, (e) 70 °C and (f) 80 °C.

direction with a small domain size (2 μm–5 μm), and the other domain had domain walls parallel to the [110] direction with a large domain size (20 μm–50 μm). With a temperature increase (to 40 °C), the small domain walls disappeared and new large domain walls parallel to the [110] direction were observed. Comparing the P and Q regions, as shown in Fig. 1(b)–(d), the non-180° domain walls changed with a temperature increase. Meanwhile, the interference colors also changed, as shown in Fig. 1(b)–(f), which may be associated with polarization rotation when the temperature increases from 40 °C to 80 °C. As shown in Fig. 1(e) and (f), the large domain walls were slightly deviated from the [110] direction, and are related to the unstable polarization state at the temperature near the ferroelectric-to-ferroelectric phase transition.

The domain structure for the [001]-oriented 0.23PIN–0.42PMN–0.35PT crystals was investigated in the temperature range of 200 °C to 205 °C, as shown in Fig. 2. Generally, the 180° domain walls are not detected by the PLM because the polarization vectors are along the same direction in the poled samples. However, the 180° polarization vectors are anti-parallel in the unpoled samples and the 180° domain wall, which shows a black-white pattern, can be detected by the PLM. Comparing Fig. 2(a) and (b), the 180° domain walls were detected at 202 °C, which is indicative of the 180° domain switch at this temperature. When the temperature was increased, the 90° and 180° domain coexisting region, marked as the S region, was broadened, as shown in Fig. 2(b)–(d). The colorful 90° domain region changed to a black-white color, as shown in Fig. 2(e) and (f); meanwhile, new 180° domain walls were found in Fig. 2(f). At 203.8 °C, part of the domain structure disappeared, which is related to the ferroelectric-to-paraelectric

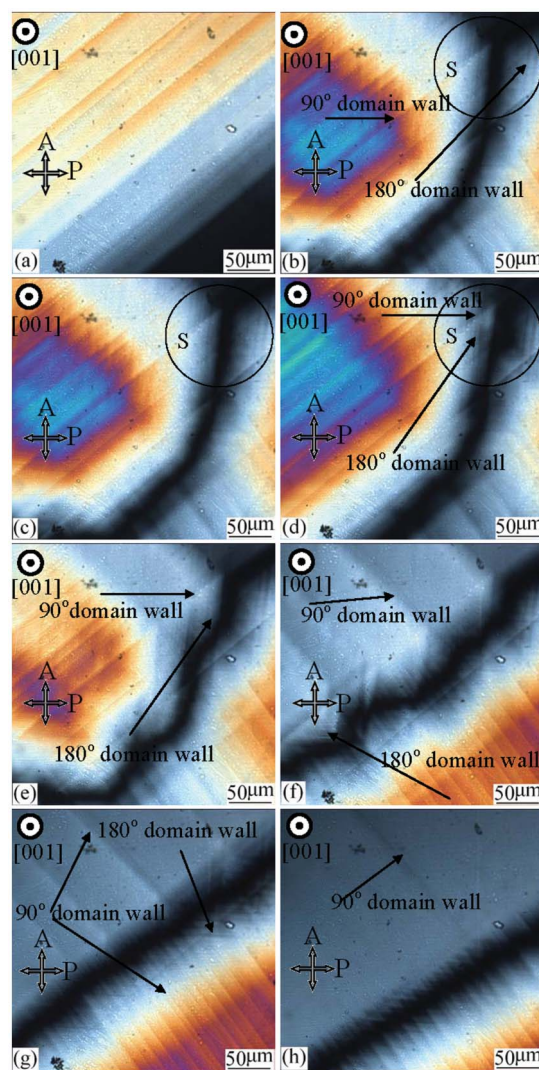


Fig. 2 The domain structures observed in the [001]-oriented PIN–PMN–PT crystals at temperatures of (a) 200 °C, (b) 202 °C, (c) 202.3 °C, (d) 202.4 °C, (e) 202.5 °C, (f) 202.6 °C, (g) 203.8 °C and (h) 204.1 °C.

phase transition, as shown in Fig. 2(g). Comparing Fig. 2(g) and (h), the number of 90° and 180° domain walls decreased when the temperature increased, and they totally disappeared at 205 °C.

Fig. 3 shows the temperature dependence of the dielectric permittivity (ϵ_r) and dielectric loss ($\tan \delta$) for the [001]-poled 0.23PIN–0.42PMN–0.35PT crystals under a zero bias field. Two dielectric permittivity peaks, peak I (~50 °C) and peak II (~207 °C), were observed in the poled samples. Peak I is the M_C -to-T phase transition, while peak II reflects the T-to-C phase transition. Two distinct dielectric loss peaks, peak I-I (~60 °C) and peak I-II (~80 °C), were observed at temperatures higher than T_{M-T} . Correspondingly, the dielectric permittivity curve shows a step-like decrease when the temperature increases from 60 °C to 80 °C. It is of particular interest that some other step-like dielectric loss anomalies were found before peak I, which correspond to the M_C -to-T phase transition. Furthermore, two novel dielectric loss peaks were detected at 201 °C and 207 °C, respectively. Moreover,

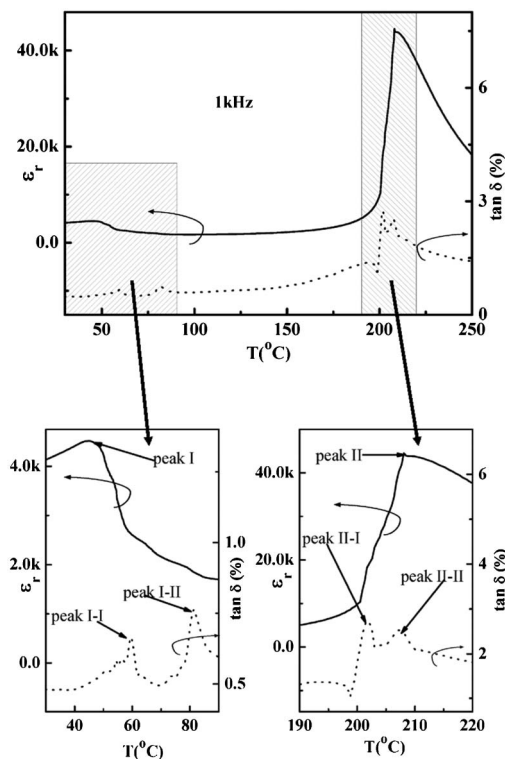


Fig. 3 The dielectric permittivity and dielectric loss measured as functions of temperature in the [001]-oriented 0.23PIN-0.42PMN-0.35PT crystals.

the dielectric permittivity curve showed a step-like increase from 201 °C to 207 °C.

Two dielectric permittivity peaks corresponding to the R-M and M-T phase transitions, were detected in the [001]-poled rhombohedral PMN-0.30PT crystals.^{8,9} Meanwhile, two dielectric loss peaks, one associated with the R-to-M_A phase transition and one associated with the M_A-to-M_C phase transition, indicating a two-step change were found near T_{R-M} .^{8,9} However, only one permittivity peak (~ 50 °C) was observed near T_{M-T} for the [001]-poled 0.23PIN-0.42PMN-0.35PT crystals, as shown in Fig. 3. Furthermore, no dielectric loss peak was observed below T_{M-T} . Based on the dielectric property analysis, the M_C phase was thought to dominate the [001]-poled 0.23PIN-0.42PMN-0.35PT crystals at room temperature. The phase transition path of R \rightarrow M_A \rightarrow M_C \rightarrow T has been confirmed in the [001]-oriented PMN-PT crystals in an electric field, based on XRD and neutron diffraction scattering research.¹⁰ In addition, the stable monoclinic phase has been reported in the [001]-poled PMN-0.30PT crystals, based on the dielectric measurements.⁸ That is to say, the polarization rotated to the [001] direction. Based on the domain observations and the dielectric behavior, the polarization rotation path was M_C-T in the [001]-oriented 0.23PIN-0.42PMN-0.35PT crystals with an electric field applied along the [001] direction.

Direct domain wall motion was observed in the [001]-poled 0.23PIN-0.42PMN-0.35PT crystals, as shown in Fig. 1 and 2. In order to confirm the factor contributing to domain wall motion, the polarization hysteresis loops were measured at increasing temperatures for the 0.23PIN-0.42PMN-0.35PT crystals. Fig. 4

shows the temperature dependent (a) remnant polarization (P_r) and (b) coercive field (E_c). The P_r and E_c were found to be 28 $\mu\text{C cm}^{-2}$ and 6.3 kV cm^{-1} in the [001]-oriented 0.23PIN-0.42PMN-0.35PT crystals at room temperature, respectively. P_r exhibited a clear drop near 40 °C, implying a significant polarization reorientation of the local domains. This is consistent with the M_C-T phase transition temperature, as shown in Fig. 3, and the evident domain wall motion in Fig. 1(a) and (b). With an increase in temperature, the polarization vector of M_C rotated to T below T_{M-T} , while some other polarization vector turned back to M_C because of the unstable polarization state. Therefore, non-180° domain wall motion was observed when the temperature increased from 40 °C to 50 °C, as shown in Fig. 1(a)-(c), which was induced by the polarization rotation. When the temperature was further increased, the polarization vector totally turned to the [001] direction due to the M_C-to-T phase transition, and clear non-180° domain wall motion could be observed at temperatures from 50 °C to 80 °C, as shown in Fig. 1(c)-(f). Based on the above analysis, it can be concluded that the domain wall motion observed in Fig. 1 was induced by polarization rotation.

In the *in-situ* domain observations as a function of temperature for the [001]-oriented 0.23PIN-0.42PMN-0.35PT crystals, the small domain structures were found to disappear at 40 °C, while domain wall motion was observed from 40 °C to 80 °C. In the dielectric measurements, a dielectric permittivity peak was detected at 50 °C, which showed a step-like decrease when the temperature increased from 60 °C to 80 °C. Furthermore, step-like dielectric loss anomalies were found from 30 °C to 50 °C, and two distinct dielectric loss peaks were observed from 60 °C to 80 °C. Comparing the results from the domain observations and dielectric properties, the change in the small domain structure was thought to be associated with the M_C-T phase transition; moreover, the domain wall motion contributed to the dielectric loss and dielectric permittivity. The non-180° domain walls contributed to the dielectric properties, which is similar to previous research.^{11,12} In the PMN-PT crystals, no additional dielectric loss peak was observed at temperatures higher than T_{M-T} , which is due to the restriction of the domain wall motion by the external dc field. In the present work, there was no more phase transition between the M_C and T phases in the PIN-PMN-PT crystals. Therefore, it is reasonable to conclude that the additional dielectric loss peaks and step-like dielectric permittivity behavior in the [001]-oriented 0.23PIN-0.42PMN-0.35PT crystals were induced by the domain wall motion, and the domain wall motion was related to the unstable polarization state through the polarization rotation.

On the other hand, it has been reported that a dielectric loss peak exists at temperatures a few degrees below T_C in ferroelectric materials such as KDP, PMN-PT and PIN,¹⁻³ which was thought to be related to domain wall motion. In this work, direct 90° domain wall motion was observed from 200 °C to 205 °C; meanwhile, an additional dielectric loss peak was found at 201 °C. It is of particular importance that the 180° domain switch was detected at 202°, as shown in Fig. 2, and the 90° domain wall motion was found to be driven by the 180° domain switching. Based on the results of the domain observations and dielectric properties, it can

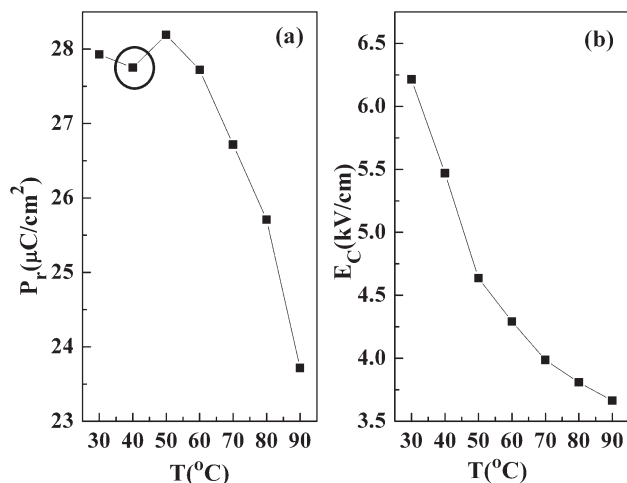


Fig. 4 The temperature-dependent (a) remnant polarization (P_r) and (b) coercive field (E_c) for the [001]-oriented 0.23PIN-0.42PMN-0.35PT crystals.

be concluded that the abnormal dielectric loss near the ferroelectric-to-paraelectric phase transition was related to the 180° domain switching and 90° domain wall motion.

Based on the domain structure observations, a 90° and 180° domain co-switching mechanism was proposed to explain the new domain observation near the ferroelectric-to-paraelectric phase transition, as given in Fig. 5. A description of the 90° and 180° domain walls in the tetragonal PIN-PMN-PT crystal is given in

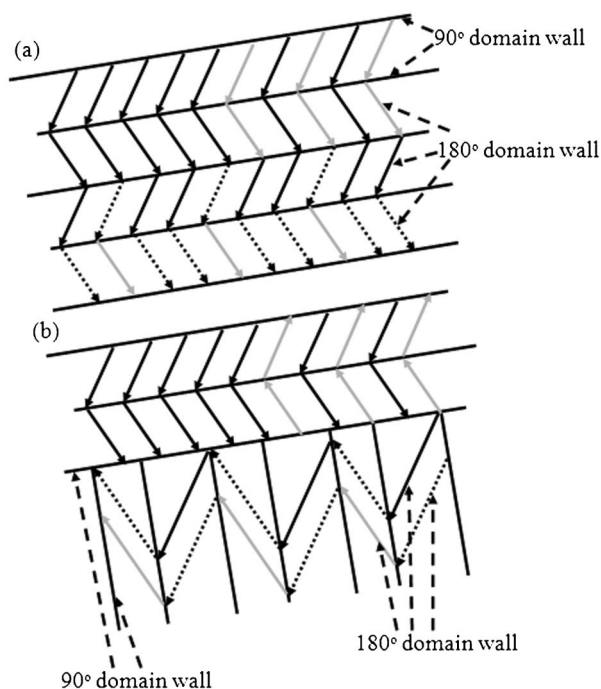


Fig. 5 Schematics of the 90° and 180° domain co-switching mechanism near the ferroelectric-to-paraelectric phase transformation. The paralleled lines represent the 90° domain walls; the solid and dotted arrows represent the 180° domain walls.

Fig. 5(a), while the change of the domain walls at temperatures below the Curie temperature is shown in Fig. 5(b). In accordance with the domain observations, the 180° domain walls were not detected by the PLM because of the parallel polarization vector. The 180° domain walls were detected by the PLM at 202°C , which indicated that part of the 180° domains reversed and became anti-parallel to the other 180° domains. Moreover, the parallel 90° domain walls shown in Fig. 2(a) were separated by 180° domain walls, and presented a vertical shape as shown in Fig. 2(b). On further increasing the temperature, the 90° domain walls rotated 90° and started to disappear. In the proposed model, partial 180° domains, marked with gray arrows, are reversed and become anti-parallel to their original direction, while other 180° domains, marked with dotted arrows, make a 90° rotation. As a result, the 90° domain walls are switched, which is due to the 180° domain wall motion. This model is consistent with the domain observations shown in Fig. 2, and can explain the new dielectric loss peak shown in Fig. 3.

Conclusions

Non- 180° domain wall motion was observed at temperatures near T_{M-T} , while 180° domain switching near 202°C (slightly lower than T_C) was observed using a PLM. The domain wall motion induced by polarization rotation was confirmed by *in-situ* domain observations and dielectric measurements, and gave rise to the two new dielectric loss peaks at temperatures a few degrees above the T_{M-T} . In addition, a 90° and 180° domain co-switching mechanism was employed to explain the domain wall motion at temperatures a few degrees below T_C .

Acknowledgements

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