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Effects of hydrostatic pressure on the dielectric response and phase transition of PMN-PT68/32 single crystal

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Abstract

Hydrostatic pressure and temperature dependence of the dielectric response and phase transition for PMN-PT68/32 single crystal have been investigated. It has been found from experimental results that hydrostatic pressure induced the phase transition from a normal ferroelectric to relaxor ferroelectric and moved T_m and T_c to lower temperature. This phenomenon is a common feature of compositionally disordered soft mode ferroelectrics.

Keywords: C. Dielectric property; PMN-PT single crystal; Hydrostatic pressure

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1. Introduction

[0 0 1]-oriented $(1 - x)$PMN-xPT ($x = 0.30$–0.35) ferroelectric single crystal near the morphotropic phase boundary (MPB) between rhombohedral and tetragonal ferroelectric phase has been known to possess high dielectric constant, piezoelectric constant, electromechanical coupling coefficient and electrically induced strain for advanced transducer and actuator applications. The PMN-PT single crystal close to MPB may be to show properties of both PMN relaxor ferroelectric and PT normal ferroelectric. Investigations have been mainly focused on the general properties under room temperature, zero electric field and zero pressure [1–3]and special properties under dc electric field [4]. Little attention has been given to hydrostatic pressure dependence of the dielectric response for this crystal. To better understand physical properties of PMN-PT single crystal under hydrostatic pressure, In this work, investigations have been performed on the dielectric response as functions of pressure and temperature at different frequency. The pressure-induced phase transition from a normal ferroelectric state (FE) to relaxor ferroelectric state (RFE) for PMN-PT68/32 single crystal is observed. This important phenomenon is similar to that for PZN-PT90.5/9.5 single crystal [5,6].

2. Experimental procedure

Reagent grade PbO, MgO, Nb_2O_5, TiO_2 powders were used as raw materials. The single crystal of the composition PMN-PT68/32 was grown by an accelerated crucible rotation technique (ACRT) and Bridgman method [3]. The single crystal was oriented along pseudocubic [0 0 1] direction by means of XRD. The silver paste was applied on the sample in size of 5 mm × 5 mm × 0.8 mm as electrodes for the dielectric properties measurements. The pressure was generated in the pressure apparatus using transformer oil as the pressure transmitting media. The pressure measurements were calculated from the resistance of a manganin wire stress gauge in the pressure cavity using a computer controlled Keithley 2000 multimeter with the accuracy better than 0.2%. To study the pressure dependence of dielectric properties at room temperature and the temperature dependence of dielectric properties under different pressure, the specimens were amounted into the pressure vessel, heated by a small resistance-heating furnace. Dielectric properties were simultaneously recorded by a HP4274 LCR meter at 0.4, 1, 10, and 100 kHz frequency.

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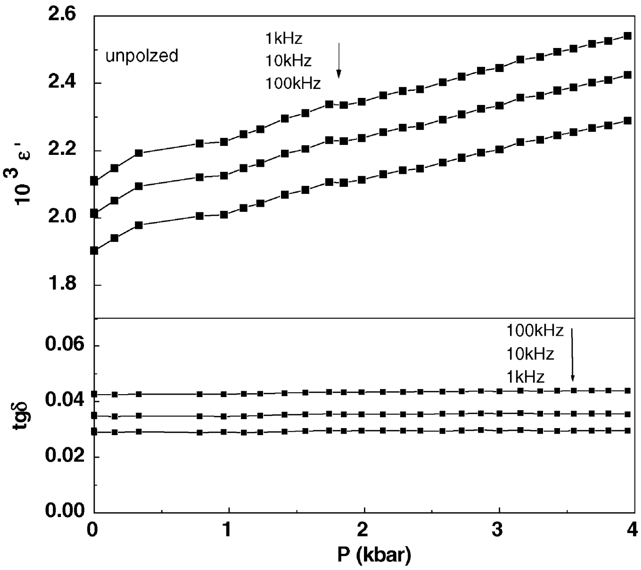

Fig. 1. Hydrostatic pressure dependence of ε' and tgδ for the crystal at room temperature.

3. Results and discussions

Fig. 1 shows the hydrostatic pressure dependence of dielectric constant ε' and loss tgδ at room temperature for [0 0 1] PMN-PT 68/32 single crystal. The dielectric constant ε' increases almost linearly with pressure, it shows strong frequency dispersion in the curve ε'–P and tgδ–P during pressure range from 1 bar to 4 kbar. The lower frequency, the bigger dielectric constant ε', the smaller loss tgδ. This dielectric frequency dispersion under hydrostatic pressure at room temperature is similar to the dielectric frequency dispersion under 1 bar for relaxor ferroelectrics. Though pressure as a variable, the pressure-induced dielectric anomalies and phase transition at room temperature were not observed.

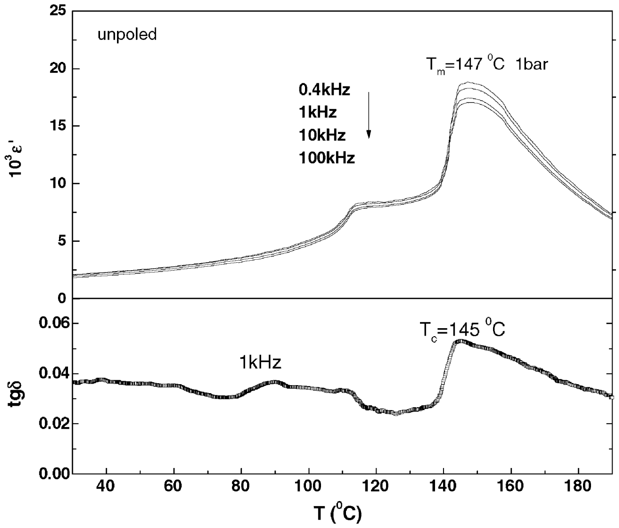

Fig. 2. Temperature dependence of ε' and tgδ for the crystal at 1 bar.

Our earlier work suggested that the PMN-PT68/32 single crystal should exhibit normal ferroelectric behavior at 1 bar [2]. Fig. 2 shows the dielectric response as a function of temperature at different frequency for unpoled crystal sample at 1 bar. It is seen that the temperature dependencies of both dielectric real ε' and loss tgδ parts at 1 bar are anomalous from tetragonal ferroelectric (FE) phase to cubic paraelectric (PE) phase. There is a remarkably sharp peak in the curve ε'–T and the phase transition is thermodynamically first order transition, thermal hysteresis is about ∼4 °C for the complex compositionally disordered mixed crystal. The curve ε'–T has a sharp peak at $T_m = 147\,^\circ$C for FE-PE phase transition at 1 kHz, T_m is the peak temperature of the maximum dielectric constant ε'_m. The curve tgδ–T exists a sharp peak at $T_c = 145\,^\circ$C, T_c is the transition temperature of the maximum dielectric loss (tgδ)$_m$. The dielectric behavior is basically like normal ferroelectrics, and the peak temperature T_m and the transition temperature T_c are essentially independent of frequency. The relatively weak frequency dispersion in the curves ε'–T at and above T_m is characteristic of some mixed perovskite ferroelectrics in PMN-PT68/32 system. The rhombohedral to tetragonal ferroelectric phase transition occurs about 80–100 °C for this crystal, There are weak shoulders in the curves ε'–T and tgδ–T about 80–100 °C, and the thermal hysteresis is about ∼12 °C, so the rhombohedral to tetragonal ferroelectric phase transition has characters of a first order phase transition, too.

As pressure increases, the dielectric responses change gradually as shown in Fig. 3. Dielectric peaks are broaden and frequency dispersion is strong step by step, and both the peak temperature T_m and the transition temperature T_c decrease. T_m decreased to 138 °C at 1 kbar and 133 °C at 4 kbar, T_c decreased to 135 °C at 1 kbar and 120 °C at 4 kbar, respectively. The frequency dispersion enhance at 4 kbar more than at 1 kbar, There is a small shoulder in the curve ε'–T

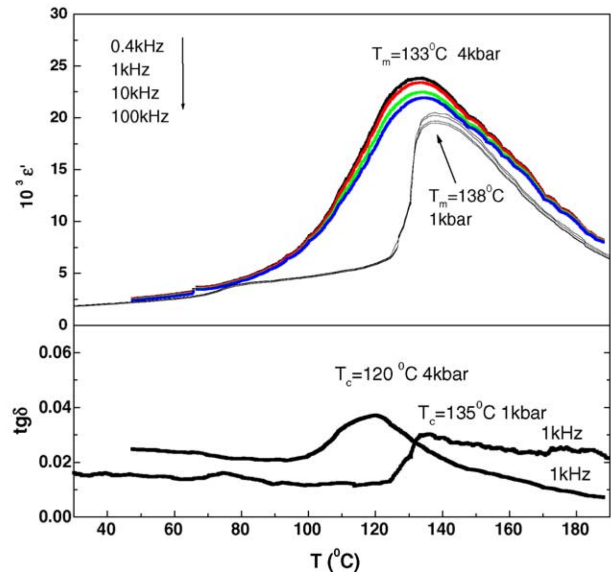

Fig. 3. Temperature dependence of ε' and tgδ for the crystal at 2 and 4 kbar.

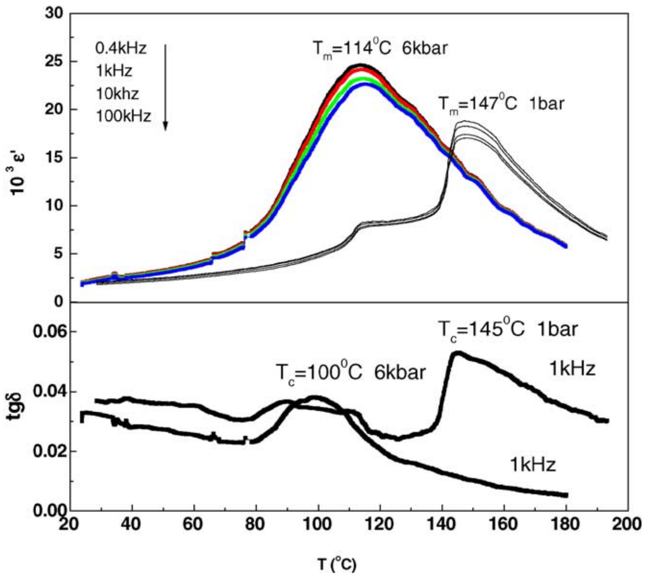

Fig. 4. Temperature dependence of ε' and tgδ for the crystal at 6 kbar and 1 bar.

at 1 kbar from the rhombohedral to tetragonal ferroelectric phase transition near ~80 °C, the shoulder is smaller than at 1 bar. Especially, there is no shoulder below 80 °C and only there are dielectric real peak at $T_{\mathrm{m}} = 133\,^\circ\mathrm{C}$ and loss peak at $T_{\mathrm{c}} = 120\,^\circ\mathrm{C}$ at 4 kbar in the curve. It is meaning that the transition from the rhombohedral to tetragonal ferroelectric phase disappear. and the transition completely vanishes at and above 6 kbar as shown in Fig. 4. Thus the rhombohedral ferroelectric phase exists at lower temperatures, yet.

Fig. 4 shows that dielectric peak is broaden further and dielectric dispersion is stronger and T_{m} decreases to 114 °C and T_{c} to 100 °C further at 6 kbar, respectively, and the rhombohedral to tetragonal ferroelectric phase transition completely vanishes at higher pressure as shown in Figs. 3 and 4. The phase transition from normal to relaxor ferroelectric state has been induced by hydrostatic pressure with increasing pressure, Also the pressure makes T_{m} and T_{c} shift to lower temperature with $\mathrm{d}T_{\mathrm{m}}/\mathrm{d}P = -5.5\,^\circ\mathrm{C/kbar}$ and $\mathrm{d}T_{\mathrm{c}}/\mathrm{d}P = -7.5\,^\circ\mathrm{C/kbar}$ averagely. This result is can be explained in terms of soft mode theory at high pressure.

Samara has proposed that the pressure-induced transition from normal to relaxor ferroelectric for PZN-PT90.5/9.5 single crystal can be explained in terms of a large decrease in the correlation length r_{c} among the polar nanodomains with increasing pressure [5,6]. It is thus clear from the above that PMN-PT68/32 single crystal also exhibits a pressure-induced phase transition from normal to relaxor ferroelectric behavior. PMN-PT68/32 system is compositional disorder, this composition disorder results in the formation of local polar clusters or nanodomains. These nanodomains are dispersed as islands in the host lattice and result from short-range correlated ionic displacements. With decreasing T at low pressures, the correlation length r_{c} increases, and the rapidly increasing r_{c} couples these nanodomaims into rapidly growing polar clusters and their Coulombic interactions enhance. Finally, these clusters spread through the whole sample and aggregate a static, cooperative long-rang ordered ferroelectric phase at $T < T_{\mathrm{m}}$. With decreasing T and at sufficiently high pressure in the EE phase at $T < T_{\mathrm{m}}$, the long-range order is destroyed and growing clusters are broken by increasing pressure, thus the correlation length r_{c} and the correlation volume decrease in a few kilobars. As the correlation length decreases continuously and the polar clusters become smaller and smaller with increasing pressure, the existence of nanodomains in the ferroelectric phase results in the appearance of frequency dispersion and relaxor behaviors. The observed dielectric relaxation and frequency dispersion enhance with increasing pressure. It is thus sufficient to induce the relaxor ferroelectric at higher pressure. This pressure-induced dielectric relaxation is more obvious and clear in physical mechanism than the temperature-induced dielectric relaxation.

4. Conclusions

The temperature dependences of dielectric response for PMN-PT68/32 single crystal under different hydrostatic pressure have been researched. Hydrostatic pressure makes T_{m} and T_{c} shift to lower temperature with $\mathrm{d}T_{\mathrm{m}}/\mathrm{d}P = -5.5\,^\circ\mathrm{C/kbar}$ and $\mathrm{d}T_{\mathrm{c}}/\mathrm{d}P = -7.5\,^\circ\mathrm{C/kbar}$ averagely and induces phase transition from normal to relaxor ferroelectric state with increasing pressure.

Acknowledgements

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