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dc-Bias Effect on Phase Transition Instabilities in PMN-PT Relaxor Ferroelectric Single Crystals

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1. Introduction

Single crystals of the solid solutions of $Pb(Mg_{1/3}Nb_{2/3})O_3$ (PMN) and prototypical relaxor ferroelectric (RFE) oxide with $PbTiO_3$ (PT), a pure ferroelectric material, have been investigated extensively due to their unusual piezoelectric and electromechanical properties [1]. Of special interest, are the compositions near the morphotropic phase boundary (MPB), where the rhombohedral *R* (3*m*) and tetragonal *T* (4*mm*) phases are separated by a small region exhibiting very high piezoelectric constant (d_{33} >2500 pC.N^{-1}), electromechanical coupling coefficient (k_{33} >94%), and hight strain values (1.7%) [1, 2]. The compositions near the MPB find extremely (with monoclinic and/or othorhombic phases) attractive applications in making large displacement actuators, high sensitivity medical ultrasonic imaging transducers, ferroelectric nonvolatile random access memories (NVRAMs) and high bit density metal-oxide-semiconductor (MOS) dynamic random access memories (DRAMs).

In perovskite relaxors, cations occoupying *B*-site have difference valencies that favor 1:2 ratio for charge conservation but due to frustration with lattice strain, *B*-site is randomnly occupied resulting in charge imbalance and hence local randomn fields preventing development of any long range polar order. In RFEs, this frustration may only be compensated by the application of an appropriate external electric field. Phenomenologically, the nanosclae polar entities, the so-called polar nano regions (PNRs) that appear at the Burns temperature (T_B) [3], well above the expected Curie temperature, are at the heart of almost all the theoretical models proposed so for to understand the physical mechanism responsible for the extraordinary dynamical properties of RFEs. Formation and thermal evolution of PNRs differentiates clearly RFE state from that of normal ferroelecrtic one even in their high temperature paraelectric phase for $T<T_B$. However, it is still far from certain that what exactly is the origin of PNRs, and do they exist reality?

To have more insight on the role of PNRs in relaxor materials, in present study we have investigated the effeccts of dc-bias on the dielectric anomalies of three kinds of PMN-PT single crystals ([001], [110] and [111]) with compositions in the MPB range.

2. Experimental

Three kinds of PMN-PT single crystals with orientations [001], [111] and [110] were grown by the Bridgeman technique. For capacitance measurements Dotite D-550 silver paste was applied on the larger face of all the crystals (~0.3 mm thick) and fired at 600 °C for 30 minutes to form good electrodes. For temperature variations, the crystal was put inside a cryostat cell THMSE 600 (Linkam) and frequency dependent capacitance and loss factor were measured by Agilent 4294A impedance analyzer. The probing ac-signal was kept to 1V in presence of different dc-bias voltages whereas temperature accuracy of THMSE 600 cryostat cell was within ±1°C with a stability of ±0.1 °C.

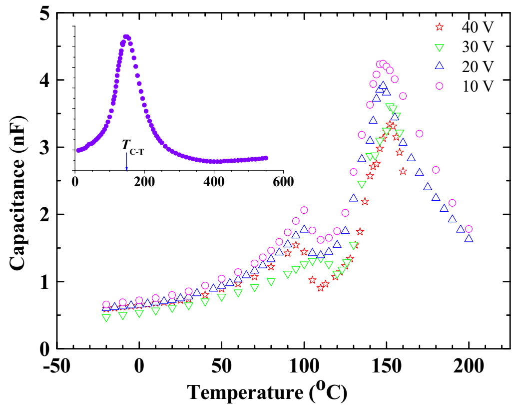

Fig. 1. Temperature dependence of capacitance of a PMN-PT [001] crystal under dc-bias measured at 1 kHz. Inset shows capacitance of the same crystal without bias voltage.

3. Results and discussion

In Fig. 1 temperature dependence of capacitance of [001]-oriented PMN-PT unpoled crystal is plotted. These data were measured by applying different dc-bias voltages upto a maximum of 40V. Inset shows data in broader temperature range at no dc-bias conditions exhibiting a large anomaly at ~150 °C related to paraelectric cubic to ferroelectric tetragonal (T_{C-T}) symmetry transformation. It can be seen that dc-bias has a substantial effect on capacitance of the crystal. With increasing dc-bias there is decrease in magnitude of capacitance at T_{C-T} with a ~4 °C shift in T_{C-T}

towards higher temperature. Another anomaly appears at ~100 °C which is moved to ~110 °C at 30V but shifts back to ~95 °C with further increase in dc-bias. Frequency dependence of capacitance at one dc-bias voltage is shown in Fig. 2 for this crystal.

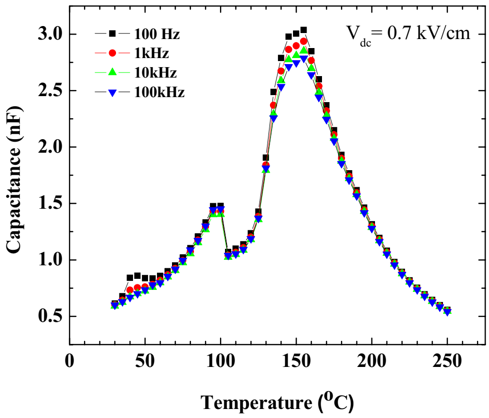

Fig. 2. Capacitance versus temperature of PMN-PT [001] crystal at some selected frequencies and bias voltage of 0.7 kV/cm.

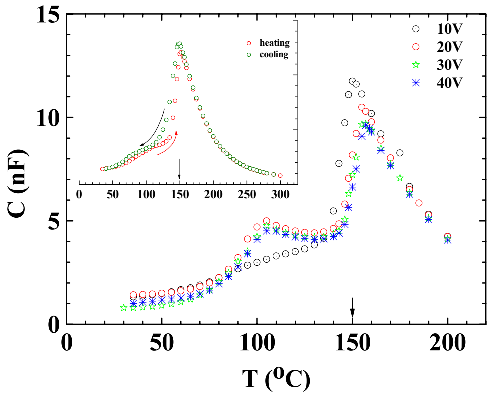

Fig. 3. Capacitance-temperature plot of PMN-PT [111] crystal under dc-bias measured at 1 kHz. Inset shows heating-cooling curves with no dc-bias.

The measured data for [111] unpoled crystal are plotted in Fig. 3. This crystal shows a clear hysteresis in heating and cooling data at both structural phase transition temperatures (inset Fig. 3) in good agreement with our previous results [4]. Under the dc-bias conditions, peak value of capacitance at $T_{C\text{-}T}$ is lowered with increasing bias voltage and transition point becomes higher as compared to zero-biased value. The most interesting phenomena is that lower phase transition anomaly at ~85 °C disappears and a rather sharper anomaly is observed at ~105 °C for bias voltage greater than 10V and remains stable with further increase in bias voltage. Further the values of capacitance well above $T_{C\text{-}T}$ show no bais dependence that means that dc-bias effect is different in ferroelectric and relaxor regimes (where PNRs are assumed to be present) in these crystals in the temperature range investigated presently.

In the rhombohedral phase polarization is directed along the [111] direction whereas it is along [001] in tetragonal phase. But for the MPB region, polarization may be rotated in (110) plane for monoclinic M_A or M_B and in (010) plane for M_C symmetry. We have discussed present results in the light of polarization rotation mechanism and co-existence of monoclinic/orthorhombic phases in the MPB region [5].

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