Large Field Property Assessment of Mn:PIN-PMN-PT Crystals for High Power Transducers

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***Abstract*—The third generation of advanced relaxor-PT piezoelectric single crystals, Mn:PIN-PMN-PT (manganese doped $Pb(In_{1/2}Nb_{1/2})O_3$-$Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$), has attracted broad attention for high power transducer applications due to their greatly reduced mechanical loss but well-maintained, extremely high electromechanical coupling coefficient. Small signal characterization of Mn:PIN-PMN-PT crystals have been extensively studied. However, systematic large signal tests of Mn:PIN-PMN-PT crystals were urgently needed for further assessing the properties for high power transducer operation. In this work, systematic large signal, quasistatic measurements have been conducted for Mn:PIN-PMN-PT crystals under variable prestress, temperature and electric field. Preliminarily optimized operation domains for high power transducer design have been revealed for [001]-poled crystals for achieving maximum acoustic power density with low hysteretic losses caused by nonlinearity and phase transitions.**

Keywords— Piezoelectric single crystal; Mn:PIN-PMN-PT; large signal measurement; high power; transducer

I. INTRODUCTION

The third generation of advanced relaxor-PT piezoelectric single crystals, Mn:PIN-PMN-PT (manganese doped $Pb(In_{1/2}Nb_{1/2})O_3$-$Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$), which were initially developed by TRS, has attracted broad attention for the next generation of high power transducers due to their greatly improved electromechanical "hardness" (low mechanical loss) with well-maintained, extremely high electromechanical coupling coefficient [1-3].

Systematic small signal characterization of Mn: PIN-PMN-PT crystals had already been done, including a full set of piezoelectric, dielectric, and elastic properties derived from small signal resonant measurements [2]. However, systematic large signal tests of Mn: PIN-PMN-PT crystals were urgently needed for further assessing the properties for high power transducer operation. As many high-power under water transducers and high-intensity focused ultrasound (HIFU) transducers may be driven under large electric field and operated under high mechanical stress and high temperature conditions [4-5], the performance of Mn: PIN-PMN-PT crystals may be well suited for them. These systems are operated over short time duration, however, a very high source level and bandwidth may be required. It is therefore important to determine not only the small signal characteristics, but also determine whether the crystals maintain the bandwidth and linear performance under a high drive field condition.

In collaboration with Material Research Institute (MRI) at the Pennsylvania State University (PSU), TRS Technologies, Inc. conducted a systematic study of the linear and nonlinear behaviors of Mn: PIN-PMN-PT crystals under large electric field. The main goal was to map out the optimal operating regimes for Mn: PIN-PMN-PT crystals for high power transducers through large signal measurements under a combination of compressive uniaxial prestress, high temperature and DC bias field. In this work, the nonlinear behavior was studied by large signal quasistatic measurement of piezoelectric and dielectric properties under mechanical prestress, electric field and temperature loadings similar to the operating loadings of high power transducers. This would provide the ranges to achieve maximum acoustic power density with low hysteretic losses caused by nonlinearity and phase transitions.

II. EXPERIMENTAL

For ferroelectric Mn:PIN-PMN-PT crystals, the polarization versus electric field (P-E loop) and strain versus electric field (S-E loop) under both bipolar and unipolar drive were measured corresponding to the external conditions, like frequency, temperature, dc bias and preload stress. From this quasistatic measurement, not only critical properties like remnant polarization (P_r), coercive field (E_C) and high field piezoelectric coefficient can be obtained, but also the electric field/temperature induced nonlinearity, the strain hysteresis and ferroelectric phase transitions can be characterized. Based on such estimation, the maximum linear and nonlinear operation range can be recommended for the applications with different tolerance on energy loss.

The high electric driving fields and the high dc bias fields were supplied by a ±30kV high voltage amplifier system (Trek model 30/20). By using a modified Sawyer-Tower circuit and linear variable differential transformer (LVDT) driven by a DSP lock-in amplifier (Stanford Research System RS830), the charge and displacement can be measured respectively. A heating bath filled with dielectric fluid (Galden HT 200) is attached to the system, which allows temperature adjustment between -85°C and 190°C.

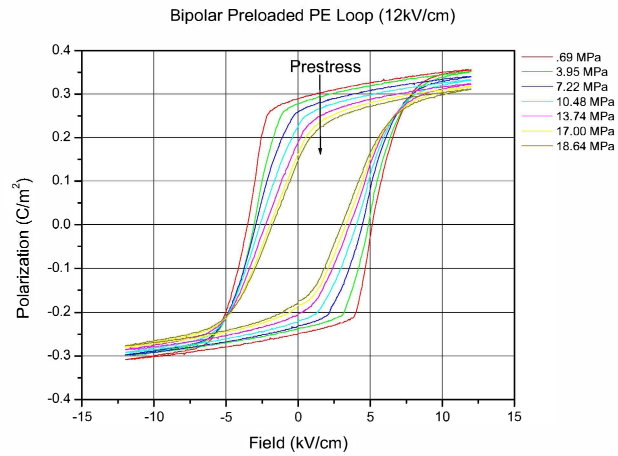
The sample fixture basically consists of two aluminum disks spaced by three steal pedestals 120° apart from one another. One of the pedestals is replaced by a crystal sample during the measurement. A spring is attached to the center of upper aluminum disk so that the uniaxial prestress can be applied to the sample. A LVDT is attached to the fixture with its body mounted on the top aluminum disk and the core screwed into the bottom aluminum disk, so that the displacement of the upper aluminum disk can be directly measured. As the crystal sample is placed right beside the LVDT core, the displacement of the crystal sample is approximately equal to that of upper aluminum disk. A crystal sample is usually sandwiched between two pieces of printed circuit boards (PCBs) with patterned copper layers contacting with the electrode on both ends of the crystal sample. Through the patterned copper layer on each of the PCB, the high AC and DC electric field can be directly applied to the crystal sample. The prestress applied to the crystal sample was monitored either indirectly by a pair of strain gauges mounted on the side surface of the loading spring or directly by a load cell placed under the crystal sample.

Thickness mode Mn: PIN-PMN-PT crystal disks with diameter of 6mm and thickness of 3mm were made for the all of the high field measurement. Crystal disks with sputtered Cr/Au electrode on both sides were poled through thickness (along [001]) under 15kV/cm at room temperature and were aged for more than 72 hours before testing.

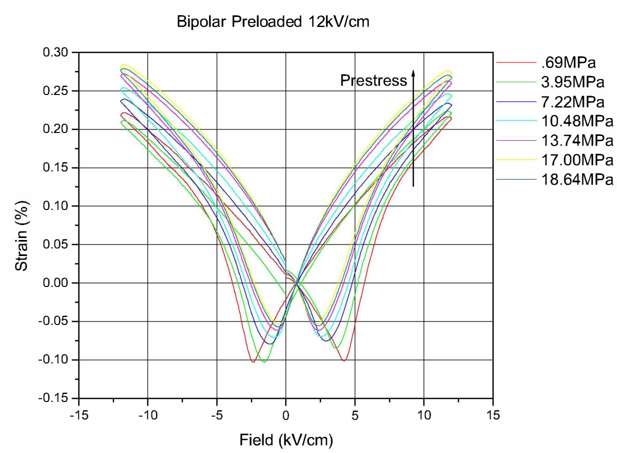
III. RESULTS AND DISCUSSION

A. *Constant Field Test under Prestress (Room temperature / no DC bias)*

The P-E and S-E loops were measured under the bipolar electric field with constant amplitude (±12kV/cm) and frequency (1Hz), but incremental prestress was applied to the crystal sample. To achieve reliable and repeatable measurement, three bipolar electric field cycles were taken under each prestress setting but only the last cycle data was recorded. Fig. 1a and 1b show the P-E loops and S-E loops respectively obtained under incremental prestress. With increasing prestress from 0.69MPa to 18.64MPa, the P-E hysteresis became skew and narrower showing clear trends of decreasing saturation polarization, remnant polarization and coercive field. With increase of prestress, S-E loops also became narrower, which means polarization switching happened at lower electric field applied opposite to the stress. Furthermore, S-E loops became more curved under high prestress, indicating the higher maximum strain was achieved through increased nonlinear loss. S-E loop measurement under unipolar electric field confirmed that the crystal produced higher strain under higher uniaxial pressure with the price of increased nonlinear loss. However, it is obvious that there was no phase transition under either unipolar or bipolar fields up to ±12kV/cm and uniaxial compressive prestress up to 18.64MPa. In above bipolar and unipolar electric field measurements, P_{sat}, P_r and E_C were extracted from the recorded P-E and S-E loops. It was shown that E_C, P_r and P_{sat} decreased almost linearly with increase of prestress, and then recovered when the prestress was withdrawn.



(a)



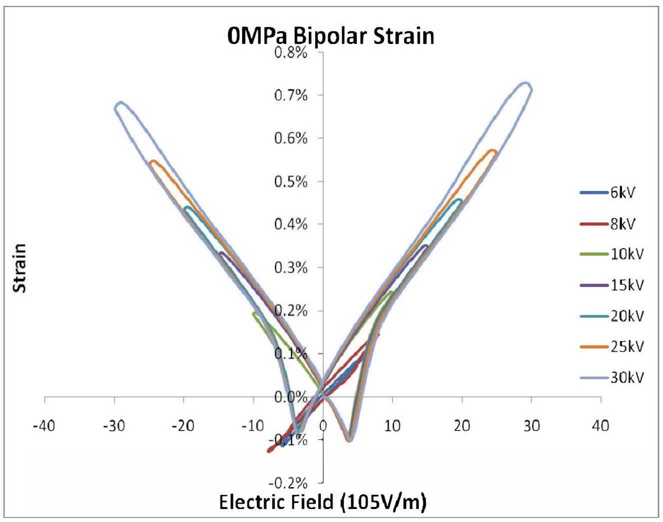
(b)

Fig. 1. The P-E loops (a) and S-E loops (b) obtained under incremental uniaxial prestress

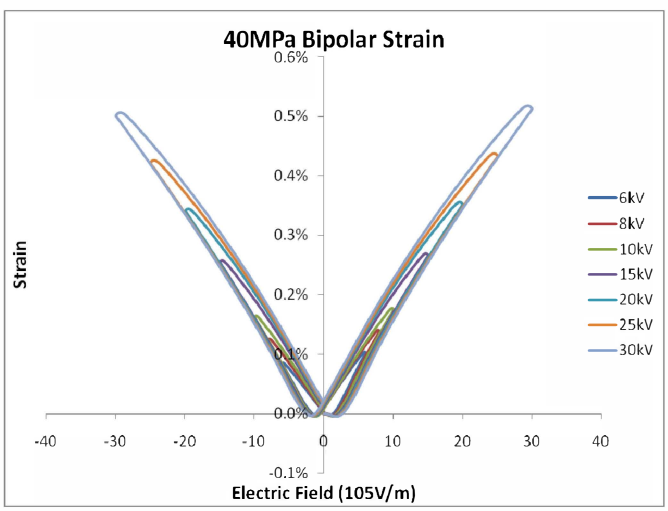
B. *Variable Field Test under Prestress (Room temperature / no DC bias)*

In this round of tests, the fixture was set for one preloading stress at a time and measurements were taken under increasing bipolar fields up to 30kV/cm. For each increment of the electrical field, three contiguous cycles of triangle wave were applied, but the measurement was only recorded on the last cycle. Prestresses used were 0, 10, 20 and 40MPa.

Fig. 2 shows the strain-electric field (S-E) loops measured under 1-30kV/cm bipolar electric field while 0MPa (a) or 40MPa (b) prestress was applied to the sample. As shown in Fig. 2a, when the bipolar electric field is equal to or smaller than 6kV/cm, the crystal sample showed linear strain-field relationship with no concern of depoling (repoling). When the bipolar electric field increased to 10kV/cm, typical "butterfly" shaped S-E loops occurred, exhibiting a maximum positive and negative strain of about 0.24% and 0.09% respectively. With further increase of driving field, the maximum positive strain kept increase with the field, while the maximum negative strain kept nearly constant. As shown in Fig 2b, the negative strain peaks became rounded under 40MPa prestress; meanwhile, the maximum negative strain reduced sharply from 0.9% at 0MPa to nearly zero at 40MPa. As a result, E_C declined sharply, which means that much lower field and less energy are required to switch the domain and depole/repole the crystal sample. As shown in Fig. 2a, without prestress the S-E loop became wider when the bipolar field reached about 25kV/cm, indicating that the rhombohedral-to-tetragonal phase transformation initiated. Fig. 2b suggests that the prestress stabilized the rhombohedral phase and increased the rhombohedral-to-tetragonal phase transformation field. This is consistent with the P-E measurement.



(a)



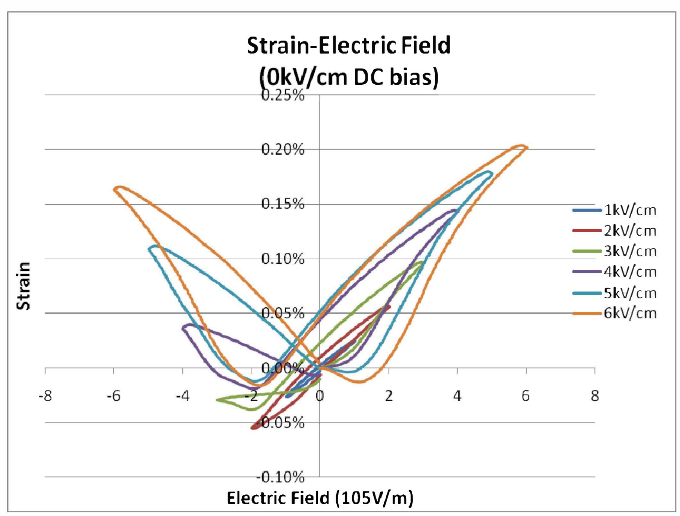
(b)

Fig. 2. Strain-electric field loops measured under 1-30kV/cm bipolar electric field while 0MPa (a) or 40MPa (b) prestress applied

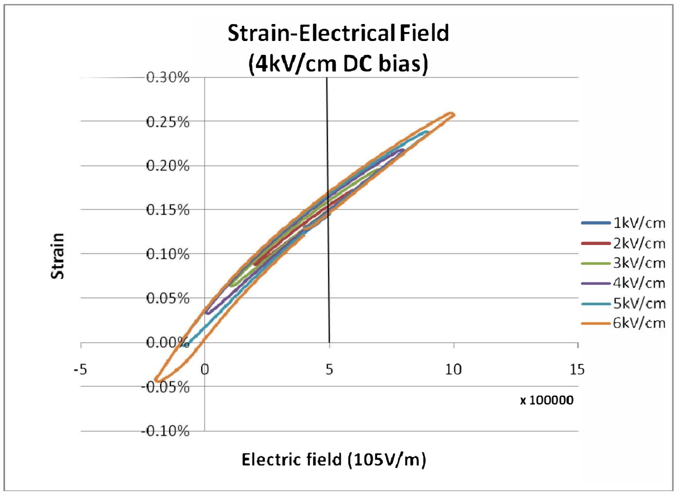
C. *High Field Test under Constant Prestress & DC bias (Room Temperature)*

All DC bias tests were performed under 10MPa uniaxial prestress at room temperature. Bipolar field tests were performed to determine charge and strain response of the sample under a 0, 2 and 4kV/cm positive DC bias (applied in the same direction of the poling field). Bipolar AC fields with triangle waveform were applied at each DC bias. Each measurement was done three times, the third being the recorded measurement, allowing for the sample to be fully poled and a normal electrical and mechanical response to be observed.

Fig. 3 shows the S-E loops with the amplitude of driven fields smaller than or equal to 6kV/cm (the measurement was done under bipolar 1-15kV/cm, 1Hz AC fields). The S-E loops measured under zero DC bias (Fig 3a), which are similar to that described in section B, are served as a baseline for analyzing the effects of DC bias. It was indicated that domain switch (depoling/repoling) process was initiated by a bipolar AC field with amplitude of 1kV/cm, which suggests that, if the crystal is driven by a low-frequency bipolar AC field under certain prestress, domain switch caused nonlinearity in the strain response may limit the amplitude of the driving field. As shown in Fig. 3b, when a positive DC bias field was applied to the crystal, the center of the electric field was shifted toward positive side. As a result of the increased amplitude of the positive field, the field-induced strain was extended as well on the positive side of the electric field. It is quite obvious that the driving electric field range, in which the crystal exhibited a linear strain response, was expanded to ±6kV/cm respectively, when a +4kV/cm DC bias was applied. The results indicated that the linear strain response range of the Mn:PIN-PMN-PT crystals under an uniaxial prestress could be recovered by application of a positive DC bias to the crystal.



(a)



(b)

Fig. 3. S-E loops measured under 10MPa prestress with no (a) and with 4kV/cm external DC bias field

D. *High Field Test under Prestress and Elevated Temperature*

Fig. 4 shows the S-E loops measured under constant prestress but elevated temperatures. The measurement procedure was similar to that was described under section B except for the temperature. When there was no prestress applied on the crystal, it was observed that the strain as well as the nonlinear loss (proportional to the hysteresis) increased with the increase of temperature. At 75°C, a field-induced phase transition occurred when the amplitude of the driving field was beyond 10kV/cm. However, as suggested by Fig. 4, the application of a compressive prestress to the crystal effectively suppressed the phase transition. When the prestress reached 20MPa, the strain response of the crystal became quite linear, and no more phase transition could be verified.

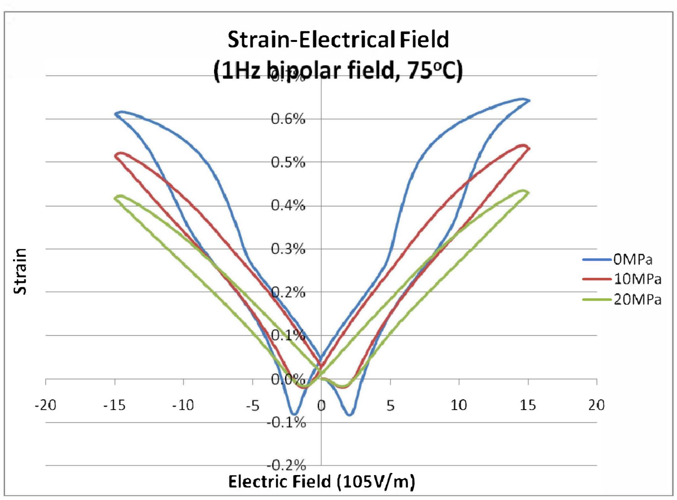


Fig. 4. S-E loops for the same sample measured at 75°C under 15kV/cm bipolar electric field

IV. CONCLUSIONS

A systematic large signal, quasistatic measurement has been conducted for [001]-poled Mn:PIN-PMN-PT crystals under variable prestress, temperature and electric field. It was proved that the uniaxial prestress can stabilize the rhombohedral phase and suppress the electric field induced phase transition; furthermore, driven by a combination of a bipolar AC field and a positive DC bias field, the crystals exhibit a broad range of linear strain and polarization response under uniaxial prestress, which may benefit the high power transducer applications. More specifically, under uniaxial prestress Mn: PIN-PMN-PT crystal kept linear strain response under large unipolar electric field; Furthermore, the bipolar driving electric field range, in which the crystal exhibited a linear strain response, was expanded to ±6kV/cm, when a +4kV/cm DC bias was applied.

ACKNOWLEDGMENT

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