

**Orientation dependence of piezoelectric properties and mechanical quality factors of  $0.27\text{Pb}(\text{In}_{1/2}\text{Nb}_{1/2})\text{O}_3\text{-}0.46\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-}0.27\text{PbTiO}_3\text{:Mn}$  single crystals**

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# Orientation dependence of piezoelectric properties and mechanical quality factors of $0.27\text{Pb}(\text{In}_{1/2}\text{Nb}_{1/2})\text{O}_3\text{-}0.46\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-}0.27\text{PbTiO}_3\text{:Mn}$ single crystals

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The complete set of material constants of single domain rhombohedral phase  $0.27\text{Pb}(\text{In}_{1/2}\text{Nb}_{1/2})\text{O}_3\text{-}0.46\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-}0.27\text{PbTiO}_3\text{:Mn}$  single crystal has been determined. The orientation dependence of piezoelectric, dielectric, and electromechanical properties was calculated based on these single domain data. The maximum piezoelectric and electromechanical properties were found to exist near the  $[001]_C$  pseudo-cubic direction. In addition, the piezoelectric properties of  $[001]_C$  poled crystals with “4R” multi-domain configuration were experimentally measured and compared with the calculated values. Only a small difference (3%) was found between experimental and theoretical values, indicating the high piezoelectric properties in the “4R” state are mainly from intrinsic contributions. The mechanical quality factors  $Q_{33}$  are significantly improved by the Mn-doping for the “4R” domain engineered crystals but almost no change for the single domain “1R” state. On the other hand,  $Q_{15}$  of both single domain and multidomain crystals were found to increase with Mn-doping, due to the internal bias induced by acceptor dopants, which clamps the domain wall motions and restricts polarization rotation. © 2013 AIP Publishing LLC.

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## I. INTRODUCTION

Relaxor-based single crystals, such as  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$  (PMN-PT), have been extensively studied in the past two decades due to their high piezoelectric coefficients ( $d_{33} > 1500\text{pC/N}$ ) and electromechanical coupling factors ( $k_{33} \sim 0.9$ ), making them excellent active materials for actuators and medical ultrasonic transducer applications.<sup>1–4</sup> However, the low rhombohedral-tetragonal phase transition temperature  $T_{R-T}$ , being in the range of  $65\text{--}95^\circ\text{C}$ ,<sup>5,6</sup> limits their usage temperature range. It was reported that the ternary ferroelectric single crystals  $\text{Pb}(\text{In}_{0.5}\text{Nb}_{0.5})\text{O}_3\text{-Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$  (PIN-PMN-PT) possessed higher Curie temperature  $T_C$  and rhombohedral to tetragonal transition temperature  $T_{R-T}$  (being on the order of  $180\text{--}220^\circ\text{C}$  and  $100\text{--}140^\circ\text{C}$ , respectively, which are about  $30^\circ\text{C}$  higher than the corresponding temperatures of PMN-PT binary system) and higher coercive field  $E_C$  (about 2 times of that for the PMN-PT crystals), showing much improved temperature stability and electric field stability, without sacrificing the high piezoelectric properties.<sup>7–9</sup>

Nevertheless, both of the PMN-PT and PIN-PMN-PT single crystals show low mechanical quality factors  $Q_m$ , preventing these materials to be used for high power transducer applications. Analogous to “hard” PZT ceramics, acceptor doping is an effective method to improve the mechanical

quality factor, which can be confirmed by the Mn modified  $\text{Pb}(\text{Zn}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$  (PZN-PT:Mn), PMN-PT:Mn, and PIN-PMN-PT:Mn single crystals.<sup>10–13</sup> However, the effect of Mn on mechanical quality factors for different domain structures and different vibration modes has not been investigated up to date. There are also no report in the literature on the full matrix of electric and mechanical properties of single-domain rhombohedral Mn doped PIN-PMN-PT (PIN-PMN-PT:Mn) single crystals, so that one could not analyze the orientation dependence of functional properties to get the optimum cut, and could not analyze the intrinsic and extrinsic contributions to the large piezoelectric properties. In addition, the complete set of self-consistent material properties is needed for practical device design of using finite element simulations.

In this work, we have measured the full set material constants for single domain  $0.27\text{PIN-}0.46\text{PMN-}0.27\text{PT:Mn}$  single crystal poled along  $[111]_C$ . Based on the single-domain data, orientation dependence of dielectric constants, piezoelectric coefficients, and electromechanical coupling factors was calculated. In addition, materials properties of  $[001]_C$  poled  $0.27\text{PIN-}0.46\text{PMN-}0.27\text{PT:Mn}$  single crystals with the engineered domain configuration “4R” were measured and compared with calculated values using single domain data, which tells us the ratios of the intrinsic and extrinsic contributions. The effect of Mn-doping on the mechanical quality factors  $Q_m$  in single domain and domain engineered single crystals was also investigated.

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## II. EXPERIMENTAL PROCEDURE

The PIN-PMN-PT:Mn single crystals were grown by the modified Bridgman method.<sup>14</sup> The as grown PIN-PMN-PT:Mn crystal boule exhibits different phases and compositions along the growth direction due to the segregation of Ti. The composition of the crystal used in this work, which was estimated by Energy Dispersive Spectrometer (EDS), is 0.27PIN-0.46PMN-0.27PT:Mn, which has rhombohedral  $3m$  symmetry and is away from the morphotropic phase boundary (MPB) with a relatively higher  $T_{R-T}$  but smaller piezoelectric coefficient.

Samples were oriented along their crystallographic directions  $[001]_C$  and  $[111]_C$  using the Laue X-ray machine with an accuracy of  $\pm 0.5^\circ$ . Vacuum sputtered gold was applied to the desired surfaces as electrodes. The  $[001]_C$  oriented samples were poled under a dc electric field of 12 kV/cm at room temperature for 10 min, while the  $[111]_C$  oriented samples were poled under the electric field of 3 kV/cm at 220 °C for 2 min, and then slowly cooled down to 50 °C with the electric field on. Single domain state can be obtained after poling along the  $[111]_C$ . The microscopic symmetry of the  $[111]_C$  poled samples is rhombohedral  $3m$ , which has 12 independent material constants: 6 elastic constants, 4 piezoelectric constants, and 2 dielectric constants. The complete set of material constants was determined by the combined ultrasonic and resonance method.<sup>15,16</sup> The resonance and anti-resonance frequencies were obtained by Agilent 4194A Impedance-phase gain analyzer. The 3 dB method was chosen for the calculation of low signal mechanical quality factors  $Q_m$ . The high field polarization and strain measurements at room temperature were performed using a modified Sawyer-Tower circuit driven by a lock-in amplifier (Stanford Research System). The temperature dependence of the dielectric properties was determined using Agilent 4284A precision LCR meter, which was connected to a computer controlled high temperature furnace.

## III. RESULTS AND DISCUSSION

### A. Dielectric and ferroelectric properties

Fig. 1 shows the dielectric constant and dielectric loss as a function of temperature for the  $[111]_C$  and  $[001]_C$  poled 0.27PIN-0.46PMN-0.27PT:Mn single crystals. For crystals in the rhombohedral phase, there are eight possible spontaneous polarization directions along  $\langle 111 \rangle_C$ . After poling along  $[111]_C$ , all polarizations align along the electric field direction through domain switching, leading to “1R” single domain state. On the other hand, for crystals poled along  $[001]_C$ , four spontaneous polarizations still remain, exhibiting “4R” engineered domain configuration. It can be seen from Fig. 1 that three dielectric anomalies exist for the  $[001]_C$  poled sample in the measured temperature range. The first one at 125 °C corresponds to the rhombohedral-tetragonal phase transition  $T_{R-T}$ . The second one at 160 °C is the depolarization temperature  $T_d$ , corresponding to the strain induced polarization reversal that causes the vanishing of the total polarization, hence, the piezoelectric property also disappears. The dielectric peak at 194 °C is the Curie

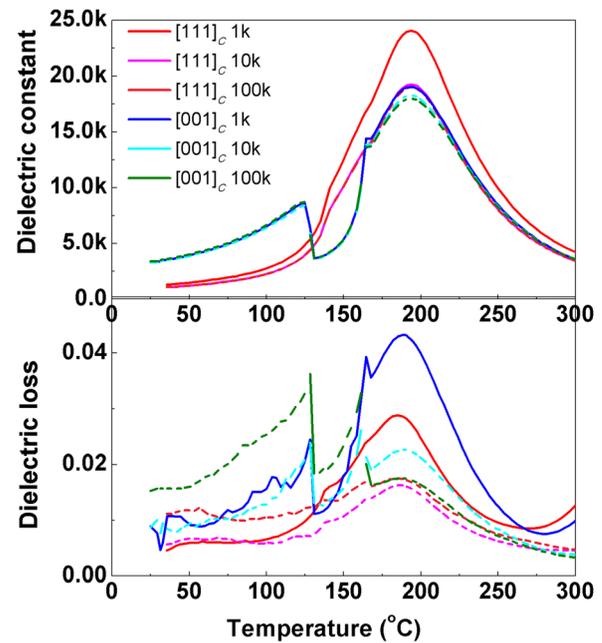


FIG. 1. Temperature dependence of dielectric constant and dielectric loss for  $[111]_C$  poled and  $[001]_C$  poled 0.27PIN-0.46PMN-0.27PT:Mn single crystals.

temperature. For the  $[111]_C$  poled single domain sample, two phase transitions were observed, i.e., the rhombohedral-tetragonal phase transition ( $T_{R-T}$ ) at 135 °C and tetragonal-cubic phase transition ( $T_C$ ) at 194 °C. The two samples were found to possess the same Curie temperature, demonstrating the same chemical composition.

It is interesting to note that the  $T_{R-T}$  for  $[111]_C$  poled sample is about 10 °C higher than that of  $[001]_C$  poled one, indicating that the ferroelectric rhombohedral to ferroelectric tetragonal phase transition temperature is orientation dependent. This is due to the fact that the free charge density on the electrode of the  $[111]_C$  poled rhombohedral crystals is much higher than that of the  $[001]_C$  poled crystals, which help stabilize the rhombohedral phase, making the rhombohedral to tetragonal phase transition to occur at higher temperature.

Fig. 2 shows the polarization hysteresis loops for  $[001]_C$  and  $[111]_C$  oriented samples measured at 1 Hz. The remnant

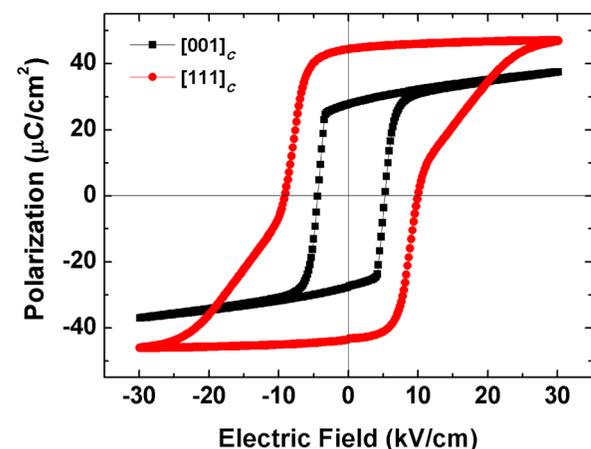


FIG. 2. Polarization hysteresis loops for 0.27PIN-0.46PMN-0.27PT:Mn single crystals measured at 30kV/cm and frequency of 1 Hz.

TABLE I. Measured and derived constants of 0.27PIN-0.46PMN-0.27PT: Mn single crystals poled along  $[111]_C$ .

Elastic stiffness constants: $c_{ij}^E$ and $c_{ij}^D$ ( $10^{10}$ N/m <sup>2</sup> )															
$c_{11}^E$	$c_{12}^E$	$c_{13}^E$	$c_{14}^E$	$c_{33}^E$	$c_{44}^E$	$c_{66}^E$	$c_{11}^D$	$c_{12}^D$	$c_{13}^D$	$c_{14}^D$	$c_{33}^D$	$c_{44}^D$	$c_{66}^D$		
19.2	8.10	6.35	1.10	18.2	1.14	5.57	20.8	7.24	5.67	-1.08	19.6	5.16	6.76		
Elastic compliance constants: $s_{ij}^E$ and $s_{ij}^D$ ( $10^{-12}$ m <sup>2</sup> /N)															
$s_{11}^E$	$s_{12}^E$	$s_{13}^E$	$s_{14}^E$	$s_{33}^E$	$s_{44}^E$	$s_{66}^E$	$s_{11}^D$	$s_{12}^D$	$s_{13}^D$	$s_{14}^D$	$s_{33}^D$	$s_{44}^D$	$s_{66}^D$		
7.74	-3.37	-1.53	-10.8	6.58	109.0	22.2	5.85	-1.80	-1.17	1.61	5.79	20.05	15.3		
Piezoelectric coefficients: $e_{iz}$ (C/m <sup>2</sup> ), $d_{iz}$ ( $10^{-12}$ C/N), $g_{iz}$ ( $10^{-3}$ Vm/N), and $h_{iz}$ ( $10^8$ V/m)															
$e_{15}$	$e_{31}$	$e_{22}$	$e_{33}$	$d_{15}$	$d_{31}$	$d_{22}$	$d_{33}$	$g_{15}$	$g_{31}$	$g_{22}$	$g_{33}$	$h_{15}$	$h_{31}$	$h_{22}$	$h_{33}$
17.5	-5.78	9.52	11.8	2111	-43	295	95	42.1	-3.76	5.88	8.26	23.0	-5.84	12.5	11.9
Dielectric constants: $\epsilon_{ij}(\epsilon_0)$ and $\beta_{ij}(10^{-4}/\epsilon_0)$							Electromechanical coupling factors								
$\epsilon_{11}^T$	$\epsilon_{33}^T$	$\epsilon_{11}^S$	$\epsilon_{33}^S$	$\beta_{11}^T$	$\beta_{33}^T$	$\beta_{11}^S$	$\beta_{33}^S$	$k_{15}$	$k_{31}$	$k_{33}$	$k_t$				
5659	1299	858	1117	1.77	7.70	11.7	8.96	0.883	0.144	0.346	0.267				

polarization  $P_r$  for the  $[001]_C$  and  $[111]_C$  oriented samples are 27.4 and 43.8  $\mu\text{C}/\text{cm}^2$ , respectively. The coercive field  $E_C$  for the  $[001]_C$  oriented sample is 4.4 kV/cm, much lower than that of the  $[111]_C$  oriented sample  $\sim 9.1$  kV/cm. In Mn modified single crystals, the acceptor cations  $\text{Mn}^{2+}$  and  $\text{Mn}^{3+}$  substitute  $\text{Ti}^{4+}$ , owing to their similar ionic radius. To maintain charge balance, oxygen vacancies were created, resulting in the formation of acceptor-oxygen defect dipoles, which occupy energetically preferred sites in the lattice, forming anisotropic nucleation centers for the formation of domains. Defect dipoles realign themselves along a preferential direction of the spontaneous polarization in a given domain, giving rise to an internal bias. The internal biases of the  $[111]_C$  and  $[001]_C$  oriented samples were found to be on the same level, being on the order of 0.44 kV/cm. It should be noted that the internal biases for  $[001]_C$  and  $[011]_C$  poled orthorhombic 0.26PIN-0.42PMN-0.32PT:Mn crystals were reported to be on the order of 0.5 kV/cm,<sup>13</sup> which is slightly higher than that of rhombohedral crystals observed by us. Therefore, it can be concluded that the level of internal bias is independent of crystallographic orientation, but only associated with the concentration of Mn. It was found that the concentration of Mn doping also shows segregation along the growth direction, so that the internal bias level generated also changes along the growth direction.

## B. Complete set of dielectric, piezoelectric, and elastic constants

The complete set of elastic, piezoelectric, and dielectric constants of “1R” single domain crystal 0.27PIN-0.46PMN-

0.27PT:Mn has been determined and is listed in Table I. The thickness shear piezoelectric coefficient and electromechanical coupling factor were found to be 2111 pC/N and 0.883, respectively, much higher than its values for longitudinal mode ( $d_{33} = 95$  pC/N and  $k_{33} = 0.346$ ). This is a typical characteristic of single domain state due to the strong anisotropy of relaxor-PT single crystals.<sup>17,18</sup>

The properties of Mn doped single domain 0.27PIN-0.46PMN-0.27PT:Mn crystal and literature available data for unmodified 0.26PIN-0.46PMN-0.28PT and 0.72PMN-0.28PT crystals are compared in Table II. These three single crystals have similar piezoelectric and dielectric properties, indicating that the introduction of Mn did not affect much on the piezoelectric performance. Compared with 0.72PMN-0.28PT single crystals, both Mn modified and unmodified PIN-PMN-PT single crystals possess much higher phase transition temperature  $T_{R-T}$  and  $T_C$ .

## C. Anisotropic behavior of piezoelectric, dielectric, and electromechanical properties

In order to determine the orientation dependence of the piezoelectric coefficient  $d_{33}^*$ , dielectric constant  $\epsilon_{33}^*$ , and electromechanical coupling factor  $k_{33}^*$ , we have performed matrix transformation in space coordinates using the single domain data in “1R” state, the results are given in Figures 3–5 in the  $(1-10)_C$  crystallographic plane. All material parameters show strong orientation dependence. It can be seen from Fig. 3 that the highest  $d_{33}^*$  value is 982 pC/N, being 57 °C canted from  $[111]_C$ . The dielectric constant  $\epsilon_{33}^*$  increases as

TABLE II. Property comparison among rhombohedral single domain 0.27PIN-0.46PMN-0.27PT:Mn, 0.26PIN-0.46PMN-0.28PT, and 0.72PMN-0.28PT single crystals.

Material	$T_C$ (°C)	$T_{R-T}$ (°C)	$d_{15}$ (pC/N)	$d_{33}$ (pC/N)	$\epsilon_{11}^T$	$\epsilon_{33}^T$	$k_{15}$	$k_{33}$
0.27PIN-0.46PMN-0.27PT:Mn <sup>a</sup>	194	135	2111	95	5659	1299	0.883	0.346
0.26PIN-0.46PMN-0.28PT <sup>15</sup>	167	123	2190	74	6286	702	0.92	0.36
0.72PMN-0.28PT <sup>6,19,b</sup>	$\sim 133$	$\sim 96$	2382	97	4983	593	0.94	0.53

<sup>a</sup>This work.<sup>b</sup>The piezoelectric and dielectric properties are from Ref. 19, while the phase transition temperatures are from Ref. 6.

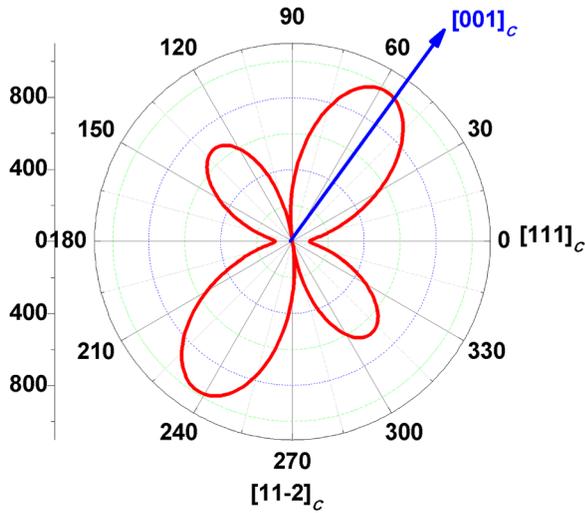


FIG. 3. Orientation dependence of  $d_{33}^*$  for single domain rhombohedral 0.27PIN-0.46PMN-0.27PT:Mn crystals in the  $(1-10)_c$  crystallographic plane.

the orientation deviated from the  $[111]_c$ , due to the larger  $\varepsilon_{11}$  in the single domain state. The highest electromechanical coupling factor  $k_{33}^*$  was found to be 0.865 at  $61^\circ$  inclined from  $[111]_c$ , as shown in Fig. 5, but the  $k_{33}^*$  value changes much slowly with orientation compared to  $d_{33}^*$ . Based on the above results, the best piezoelectric and electromechanical properties can be obtained near the  $[001]_c$ , which is  $54.7^\circ$  canted from  $[111]_c$ .

Using the single domain data, we can calculate the piezoelectric coefficients, elastic constants, and electromechanical coupling factors of  $[001]_c$  poled crystals using coordinate transformation. Neglecting domain wall contributions, the effective properties in the “4R” domain state can be obtained from the data of single domain rhombohedral state

$$d_{33}^{AR} = \frac{2\sqrt{3}}{9}(d_{31} + d_{15}) + \frac{\sqrt{3}}{9}d_{33} + \frac{2\sqrt{6}}{9}d_{22}, \quad (1a)$$

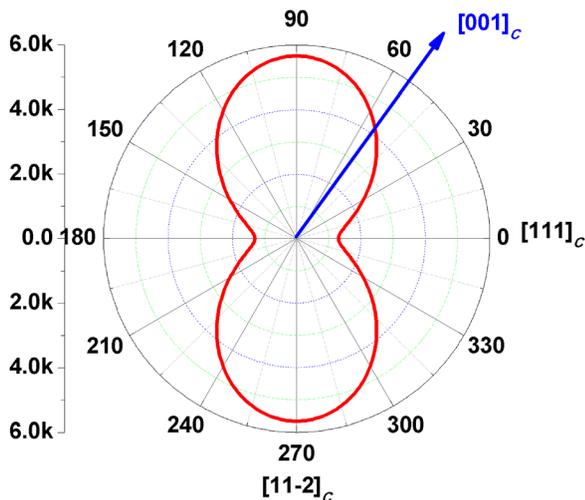


FIG. 4. Orientation dependence of  $d_{33}^{AR}$  for single domain rhombohedral 0.27PIN-0.46PMN-0.27PT:Mn crystals in the  $(1-10)_c$  crystallographic plane.

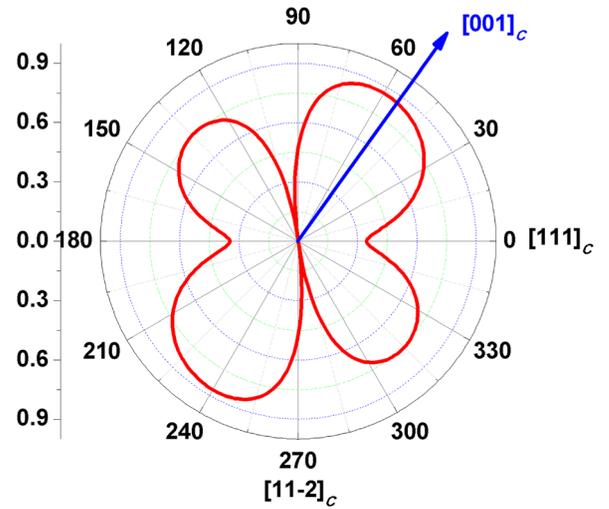


FIG. 5. Orientation dependence of  $k_{33}^*$  for single domain rhombohedral 0.27PIN-0.46PMN-0.27PT:Mn crystals in the  $(1-10)_c$  crystallographic plane.

$$d_{31}^{AR} = \frac{\sqrt{3}}{9}(2 * d_{31} + d_{33}) - \frac{\sqrt{6}}{9}d_{22} - \frac{\sqrt{3}}{9}d_{15}, \quad (1b)$$

$$\varepsilon_{33}^{AR} = \frac{2}{3}\varepsilon_{11} + \frac{1}{3}\varepsilon_{33}, \quad (1c)$$

$$s_{11}^{AR} = \frac{1}{9}(4s_{11} + 4s_{13} + s_{33} + 2s_{44} - 4\sqrt{2}s_{14}), \quad (1d)$$

$$s_{33}^{AR} = \frac{1}{9}(4s_{11} + 4s_{13} + s_{33} + 2s_{44} - 4\sqrt{2}s_{14}). \quad (1e)$$

The electromechanical coupling factors can be calculated according to the formula

$$k_{ij}^{AR} = \frac{d_{ij}}{\sqrt{\varepsilon_{ii}^{TAR} s_{jj}^{EAR}}}, \quad (2)$$

where the material constants on the right hand side are the single domain data and the parameters on the left hand side, such as  $d_{33}^{AR}$ ,  $d_{31}^{AR}$ , etc., are predicted data for the “4R” domain engineered crystals.

The corresponding material constants of  $[001]_c$  poled single crystals with the same composition are measured for comparison, as listed in Table III. It can be seen that the calculated values of the longitudinal and lateral mode are very close to the measured results, indicating that the extrinsic contributions from the domain wall motions are minimal, the high piezoelectric and electromechanical properties of “4R” domain engineered crystals are dominated by the intrinsic contributions of orientation effect. From the difference of the

TABLE III. Calculated and measured materials constants for “4R” domain engineered 0.27PIN-0.46PMN-0.27PT:Mn single crystals.

	$d_{33}$ pC/N	$d_{31}$ pC/N	$\varepsilon_{33}^T$	$s_{11}^E$	$s_{33}^E$	$k_{33}$	$k_{31}$
Calculated	975	-485	4205	34.5	34.5	0.860	0.428
Measured	1009	-481	3315	38.5	45.1	0.877	0.453

measured and calculated  $d_{33}$ , it can be estimated that the extrinsic contributions to the longitudinal piezoelectric property are about 3.3%, consistent with the value determined by the Rayleigh analysis for compositions away from the MPB.<sup>20</sup>

#### D. Mechanical quality factors of single domain and domain-engineered 0.27PIN-0.46PMN-0.27PT:Mn crystals

As mentioned above, the introduction of Mn leads to an internal bias, which clamps the domain wall motions, stabilizes domains, and restricts polarization rotation, therefore, high mechanical quality factors can be expected. Table IV summarized the mechanical quality factors of different vibration modes for 0.27PIN-0.46PMN-0.27PT:Mn single crystals with “1R” and “4R” domain configurations. From Table IV, it can be seen that  $Q_{15}$  is much lower than  $Q_{33}$  in the single domain state, indicating that the rotation of spontaneous polarization induced by the vertical electrical field is the dominant factor for the low quality factor of the thickness shear mode.

The corresponding  $Q_m$  values for some relaxor-PT crystals with different domain configurations are listed in Table IV for comparison. For single domain crystals 0.70PMT-0.30PT and 0.27PIN-0.46PMN-0.27PT:Mn with domain state of “1R” and orthorhombic 0.26PIN-0.42PMN-0.32PT:Mn with “1O” configuration, the  $Q_{33}$  values are all >1000, independent of composition, phase structure, and orientation. For domain engineered crystals, the longitudinal mechanical quality factor was increased significantly by the introduction of acceptor dopants. The  $Q_{33}$  of “4R” domain structure without Mn doping is on the order of 100–200, and increased to 650 after Mn doping. The same phenomenon was observed also in the “2R” domain structure,  $Q_{33}$  is 500 without Mn and becomes more than 1000 with Mn doping. Therefore, it can be concluded that the longitudinal mechanical quality factors of single domain crystals are not benefited by the introduction of acceptor dopants, due to the fact that there are no polarization rotation involved in the mode. On the contrary, for domain-engineered crystals, the acceptor dopants can help improve the longitudinal mechanical quality factor  $Q_{33}$ , due to the fact that the internal bias restricts

polarization rotation and clamps the domain wall motions. For the thickness shear mode quality factor  $Q_{15}$ , for both single domain crystals and domain-engineered crystals with “4R” and “2R” structures, Mn doping produced significant improvement.

In general, the mechanical loss (inverse of  $Q_m$ ) arises from two parts: rotation of the polarization and motions of domain walls. In single domain crystals, no domain wall exists. Therefore, high  $Q_{33}$  can be obtained due to the lack of E-field induced polarization rotation and domain wall motions. However, the thickness shear mechanical quality factor  $Q_{15}$  can be improved in the acceptor doped crystals due to the fact that the internal bias restricts polarization rotation. For domain engineered single crystals, the internal bias pins the motions of domain walls and also restricts polarization rotation. As a result, the mechanical quality factor  $Q_{33}$  in multidomain configurations is greatly improved.

#### IV. SUMMARY AND CONCLUSIONS

In summary, the electrical properties of rhombohedral 0.27PIN-0.46PMN-0.27PT:Mn single crystals with relatively high ferroelectric phase transition temperature  $T_{R-T}$  (around 125 °C) have been studied. The complete set of material constants was determined and the orientation dependence of longitudinal dielectric, piezoelectric, and electromechanical coupling properties were calculated based on the measured single domain data. Theoretical calculations based on single domain data predicted the appearance of maximum  $d_{33}$  and  $k_{33}$  in the direction 57° and 61° from that of [111]<sub>C</sub>, both directions are very close to [001]<sub>C</sub>, which is 54.7° canted from [111]<sub>C</sub>. The piezoelectric, dielectric, and elastic properties along [001]<sub>C</sub> direction were also directly measured and compared with the calculated values based on coordinate transformation. The results showed that the high longitudinal and lateral piezoelectric properties are associated with the strong anisotropic characteristic of the single domain state, with minimal extrinsic contributions. The magnitude of internal bias produced by the Mn-doping was found to be independent of crystalline orientation, but only depends on the Mn concentration. The mechanical quality factors  $Q_{33}$  and  $Q_{15}$  of single domain “1R” state and “4R” domain engineered state were determined and compared with undoped crystals. It was found that the introduction of Mn significantly improved the longitudinal quality factor  $Q_{33}$  for the domain engineered crystals but showed little effect on single domain crystals.

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TABLE IV. Comparison of mechanical quality factors for some PT-based relaxor crystals with different domain structures.

Material	Domain structure	$Q_{33}$	$Q_{31}$	$Q_{15}$
0.27PIN-0.46PMN-0.27PT:Mn (this work)	4R	650	200	450
	1R (single domain)	1200	350	200
PIN-PMN-PT <sup>21,22,a</sup>	4R	160	150	100
	2R	500	210	40
PMN-PT30 <sup>22,23</sup>	4R	120		80
	1R (single domain)	1130		20
PIN-PMN-PT:Mn <sup>12,a</sup>	2R	1000	260	130
0.26PIN-0.42PMN-0.32PT:Mn <sup>13</sup>	1O (single domain)	1230	490	70

<sup>a</sup>The exact composition of the crystal was not provide.

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