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Complete set of material properties of $[011]_c$ poled $0.24Pb(In_{1/2}Nb_{1/2})O_3$–$0.46Pb(Mg_{1/3}Nb_{2/3})O_3$–$0.30PbTiO_3$ single crystal

Enwei Sun [a,b], Wenwu Cao [a,b,*], Pengdi Han [c]

[a] Department of Physics, Harbin Institute of Technology, Harbin 150080, China
[b] Materials Research Institute, The Pennsylvania State University, University Park, PA 16802, USA
[c] H. C. Materials Corp., 479 Quadrangle Dr., Suite E Bolingbrook, IL 60440, USA

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ABSTRACT

A complete set of elastic, piezoelectric, and dielectric constants of $[011]_c$ poled multidomain $0.24Pb(In_{1/2}Nb_{1/2})O_3$–$0.46Pb(Mg_{1/3}Nb_{2/3})O_3$–$0.30PbTiO_3$ ternary single crystal has been determined using resonance and ultrasonic methods and the temperature dependence of the dielectric permittivity has been measured at 3 different frequencies. The experimental results revealed that this $[011]_c$ poled ternary single crystal has very large transverse piezoelectric coefficient $d_{32} = -1693$ pC/N, transverse dielectric constant $\varepsilon_{11}/\varepsilon_0 \sim 7400$ and a high electromechanical coupling factor $k_{32} \sim 90\%$. In addition, its coercive field is 2 times of that of the corresponding binary $0.69Pb(Mg_{1/3}Nb_{2/3})O_3$–$0.31PbTiO_3$ single system with much better temperature stability. Therefore, the crystal is an excellent candidate for transverse mode electromechanical devices.

1. Introduction

Recently, ternary relaxor-based ferroelectric single crystals xPb$(In_{1/2}Nb_{1/2})O_3$–$(1\text{-}x\text{-}y)Pb(Mg_{1/3}Nb_{2/3})O_3$–$y$PbTiO$_3$ (PIN-PMN-PT) with compositions near the morphotropic phase boundary (MPB) have attracted a growing attention due to their comparable piezoelectric and electromechanical properties to that of $(1\text{-}x)Pb(Mg_{1/3}Nb_{2/3})O_3$–$x$PbTiO$_3$ (PMN-PT) binary single crystals but with 2–3 times higher coercive field, and about 30 °C higher rhombohedral-tetragonal phase transition temperature (T_{R-T} ~100–120 °C) [1–7].

The $[001]_c$ poled 0.27PIN–0.40PMN–0.33PT multi-domain single crystal with MPB composition shows superior longitudinal piezoelectric strain constant ($d_{33} = 2742$ pC/N) and very high electromechanical coupling factor ($k_{33} = 0.95$) [8], while the $[111]_c$ poled rhombohedral 0.26PIN–0.46PMN–0.28PT single crystal with single-domain structure shows excellent shear piezoelectric strain coefficient ($d_{15} = 2190$ pC/N) and shear electromechanical coupling factor ($k_{15} = 0.92$) [9]. In addition, $[011]_c$ poled rhombohedral 0.26PIN–0.42PMN–0.32PT domain-engineered single crystal has been demonstrated to possess both large transverse and shear piezoelectric coefficients ($d_{32} = -1781$ pC/N and $d_{15} = 3354$ pC/N) [10]. The large d_{15} of $[011]_c$ poled case is practical more useful than that of $[111]_c$ poled crystals because multi-domain state is much more stable than single-domain state, which needs a dc bias to stabilize [11]. However, the depoling temperature of $[011]_c$ poled 0.26PIN–0.42PMN–0.32PT ($T_{R-O} = 93$ °C) is only 16 °C higher than that of $[011]_c$ poled PMN–0.31PT ($T_{R-O} = 77$ °C), which still needs improvement [10]. On the other hand, the depoling temperature of $[011]_c$ poled 0.26PIN–0.46PMN–0.28PT ($T_{R-T} = 120$ °C) can be 43 °C higher than that of PMN–0.31PT, but its piezoelectric and electromechanical coupling properties are much lower than that of 0.26PIN–0.42PMN–0.32PT due to its composition too far away from the MPB [10]. Therefore, we focus our attention to the $0.24Pb(In_{1/2}Nb_{1/2})O_3$–$0.46Pb(Mg_{1/3}Nb_{2/3})O_3$–$0.30PbTiO_3$ (0.24PIN–0.46PMN–0.30PT) system with the PT composition only slightly away from the MPB on the rhombohedral phase side, which have better temperature stability and good electromechanical properties [6,7].

In this paper, we report a complete set of elastic, piezoelectric, and dielectric constants of $[011]_c$ poled 0.24PIN–0.46PMN–0.30PT single crystal obtained by using combined resonance and ultrasonic methods. In addition, the temperature dependence of the dielectric constant and dielectric loss has been measured at frequencies of 0.1, 1, and 10 kHz to demonstrate the residue relaxor behavior.

2. Experimental

The 0.24PIN–0.46PMN–0.30PT single crystal used in this work was supplied by HC Materials Corp. (Bolingbrook, IL). The crystal was grown by the modified Bridgman method, and the crystal structure is rhombohedral with $3m$ symmetry at room temperature. The as-grown crystal was oriented by the Laue machine with an accuracy of ±0.5°. Each sample was cut and polished into a parallelepiped with three pairs of parallel surfaces along $[0\bar{1}1]_c$, $[100]_c$, and $[011]_c$, respectively. The samples were sputtered with gold electrodes on the

* Corresponding author at: 164 Materials Research Lab. The Pennsylvania State University, University Park, PA 16802, USA. Tel.: +1 814 865 4101; fax: +1 814 865 2326.
E-mail address: dzk@psu.edu (W. Cao).

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$[011]_c$ and $[0\bar{1}\bar{1}]_c$ surfaces, and poled at 10 kV/cm in silicone oil at room temperature for 30 min. Each sample was fully poled and shows a stable d_{33} when measured by using a ZJ-2 piezo d_{33} meter. The multidomain single crystal shows macroscopically orthorhombic $mm2$ symmetry after being poled along $[011]_c$, which has 17 independent material constants to be determined: 9 elastic, 5 piezoelectric, and 3 dielectric constants.

The combined resonance and ultrasonic methods has been used to get a self-consistency matrix data set. The dimensions and geometries of samples in resonance measurements were specified by the IEEE standards on piezoelectricity. A 3 mm cube sample with the face orientations of $[0\bar{1}1]_c \times [100]_c \times [011]_c$ was used for ultrasonic measurements. The resonance and anti-resonance frequencies were obtained by an HP 4194A impedance-phase gain analyzer. In the ultrasonic measurements, a 15 MHz longitudinal wave transducer (Ultran Laboratories, Inc.) and a 20 MHz shear wave transducer (Panametrics Com.) were used for the ultrasonic pulse-echo measurements. The transducers were excited by a 200 MHz pulser/receiver (Panametrics Com.) and the time of flight between echoes was measured using a Tektronix 460A digital oscilloscope. The phase velocities of the longitudinal and shear waves, which are used to calculate the elastic constants, were measured along three pure mode directions, i.e., $[0\bar{1}1]_c$, $[100]_c$ and $[011]_c$, respectively.

The dielectric properties of $[011]_c$ poled 0.24PIN–0.46PMN–0.30PT single crystal were measured as functions of temperature at different frequencies using an automated dielectric measurement system. The system consists of an HP 4284A precision LCR meter, a temperature chamber and a computer. The sample was heated from 30 °C to 250 °C at a rate of 2 °C/min and the dielectric constant was calculated from the measured capacitance based on the parallel capacitance approximation, and the dielectric loss was obtained from the same measurement.

3. Results and discussions

A complete set of elastic, piezoelectric and dielectric constants of $[011]_c$ poled 0.24PIN–0.46PMN–0.30PT single crystal has been determined from these measurements and given in Table 1. In order to investigate the composition fluctuation in the big crystal plate (~7 cm in diameter and 0.5 mm in thickness) of 0.24PIN–0.46PMN–0.30PT, three samples for each kind of vibrators were fabricated and measured. All measured constants from different samples are very consistent, which shows high-homogeneity due to the fact that the crystal boule of 0.24PIN–0.46PMN–0.30PT was grown along $[011]_c$ while the composition variation is along the growth direction, not on the growth plane.

From Table 1, we can see that the piezoelectric coefficients and electromechanical coupling coefficients of $[011]_c$ poled 0.24PIN–0.46PMN–0.30PT single crystal are much better than that of 0.26PIN–0.46PMN–0.28PT, but slightly lower than that of 0.26PIN–0.42PMN–0.32PT reported in Ref. [10]. However, the dielectric constant of 0.24PIN–0.46PMN–0.30PT single crystal is larger than that of 0.26PIN–0.42PMN–0.32PT single crystal. This is because the 0.24PIN–0.46PMN–0.30PT single crystal has higher $Pb(Mg_{1/3}Nb_{2/3})O_3$ (PMN) content than that of 0.26PIN–0.42PMN–0.32PT single crystal. It is known that PMN is a typical relaxor ferroelectric material and has extremely high dielectric constant (~9000) at room temperature [12]. Therefore, the PIN–PMN–PT ternary system shows good dielectric tunablity by adjusting the PMN content.

The coercive field of the 0.24PIN–0.46PMN–0.30PT was determined to be 5.3 kV/cm according to the polarization switching voltage level, which is comparable to that of 0.26PIN–0.42PMN–0.32PT (5.5 kV/cm), but more than two times of that of binary PMN–0.31PT (2.6 kV/cm) [10].

Fig. 1 shows the temperature dependence of the dielectric constant ($\varepsilon_{33}/\varepsilon_0$) and dielectric loss $\tan\delta$ at three different frequencies 0.1, 1, and 10 kHz, respectively, for $[011]_c$ poled 0.24PIN–0.46PMN–0.30PT single crystal. The single crystal shows typical relaxor behavior with peak shift as the frequency changes. The Curie temperature of the 0.24PIN–0.46PMN–0.30PT single crystal is 168 °C (at 1 kHz, the same below), similar to that of 0.26PIN–0.46PMN–0.28PT (165 °C) but lower than that of 0.26PIN–0.42PMN–0.32PT (192 °C) [10]. On the other hand, the rhombohedral–orthorhombic phase transition temperature T_{R-O} of 0.24PIN–0.46PMN–0.30PT is 105 °C, which is 12 °C higher than that of 0.26PIN–0.42PMN–0.32PT (T_{R-O} = 93 °C). The orthorhombic–tetragonal phase transition temperature T_{O-T} of 0.24PIN–0.46PMN–0.30PT is 113 °C, which is lower than the T_{R-T} of 0.26PIN–0.46PMN–0.28PT (120 °C) and comparable with that of 0.26PIN–0.42PMN–0.32PT (T_{O-T} = 118 °C) [10]. From these comparison, we can conclude that the $[011]_c$ poled 0.24PIN–0.46PMN–0.30PT has more application potential for devices due to its higher depoling

Table 1
Measured and derived material constants of $[011]_c$ poled 0.24PIN–0.46PMN–0.30PT multi-domain single crystal. [Directly measured constants are denoted by star (*).]

Elastic stiffness constants: c_{ij}^E and c_{ij}^D (10^{10} N/m^2)											
c_{11}^{E*}	c_{12}^E	c_{13}^E	c_{22}^{E*}	c_{23}^E	c_{33}^E	c_{44}^{E*}	c_{55}^{E*}	c_{66}^{E*}			
20.86	12.59	7.04	12.96	11.70	15.54	6.50	0.59	4.51			
c_{11}^D	c_{12}^D	c_{13}^D	c_{22}^D	c_{23}^D	c_{33}^{D*}	c_{44}^{D*}	c_{55}^{D*}	c_{66}^D			
21.00	12.13	7.78	14.49	9.22	19.49	7.17	4.57	4.51			
Elastic compliance constants: s_{ij}^E and s_{ij}^D (10^{-12} m^2/N)											
s_{11}^{E*}	s_{12}^E	s_{13}^E	s_{22}^{E*}	s_{23}^E	s_{33}^E	s_{44}^E	s_{55}^E	s_{66}^E			
20.56	−35.84	17.96	87.43	−49.47	35.56	15.39	169.49	22.17			
s_{11}^D	s_{12}^D	s_{13}^D	s_{22}^D	s_{23}^D	s_{33}^{D*}	s_{44}^D	s_{55}^D	s_{66}^D			
9.29	−7.56	0.12	16.50	−4.73	7.39	13.95	21.88	22.17			
Piezoelectric coefficients: $e_{i\lambda}$ (C/m^2), $d_{i\lambda}$ (10^{-12} C/N), $g_{i\lambda}$ (10^{-3} Vm/N), and $h_{i\lambda}$ (10^8 V/m)											
e_{15}	e_{24}	e_{31}	e_{32}	e_{33}	d_{15}	d_{24}	d_{31}^*	d_{32}^*	d_{33}^*		
18.42	9.23	2.84	−9.48	15.41	3122	142	675	−1693	1068		
g_{15}	g_{24}	g_{31}	g_{32}	g_{33}	h_{15}	h_{24}	h_{31}	h_{32}	h_{33}		
47.29	10.05	16.68	−41.82	26.38	21.61	7.21	4.70	−15.65	25.45		
Dielectric constants: ε_{ij} (ε_0) and β ($10^{-4}/\varepsilon_0$)											
ε_{11}^{S*}	ε_{22}^{S*}	ε_{33}^{S*}	ε_{11}^{T*}	ε_{22}^{T*}	ε_{33}^{T*}	β_{11}^S	β_{22}^S	β_{33}^S	β_{11}^T	β_{22}^T	β_{33}^T
963	1447	684	7459	1596	4574	10.38	6.91	14.62	1.34	6.27	2.19
Electromechanical coupling factors k_{ij} and density											
k_{15}	k_{24}	k_{31}^*	k_{32}^*	k_{33}^*	k_t^*	Density (kg/m^3)					
0.93	0.31	0.74	0.90	0.89	0.45	8161					

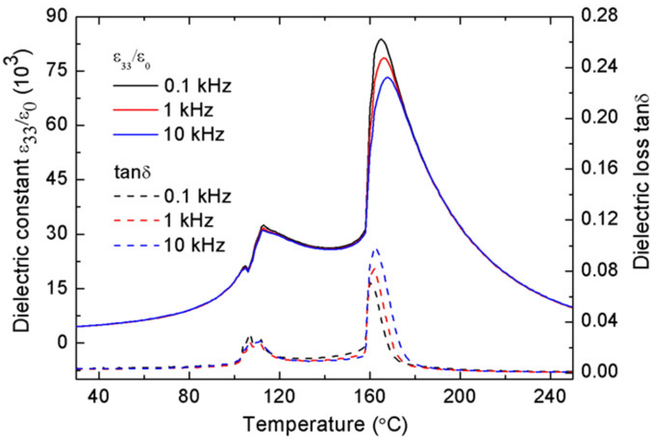

Fig. 1. Temperature dependence of dielectric constant $\varepsilon_{33}/\varepsilon_0$ and dielectric loss tan δ for 0.24PIN–0.46PMN–0.30PT single crystals in the poling direction $[011]_c$ at frequencies of 0.1, 1, and 10 kHz.

temperature, which is 12 °C and 28 °C higher than $[011]_c$ poled 0.26PIN–0.42PMN–0.32PT and PMN-0.31PT, respectively, making it more temperature stable.

4. Summary and conclusions

The elastic, piezoelectric, and dielectric constants of ternary relaxor-based ferroelectric single crystal 0.24PIN–0.46PMN–0.30PT poled along $[011]_c$ have been measured by using combined resonance and ultrasonic methods. The dielectric properties clearly demonstrated the significance of PMN content in controlling the dielectric behavior of the PIN-PMN-PT ternary single crystal system. In addition, the $[011]_c$ poled 0.24PIN–0.46PMN–0.30PT has about 30 °C higher depoling temperature than the corresponding PMN-PT binary single crystal and a factor of 2 increase in the coercive field, showing a much wider working temperature range and higher driving field level edurance.

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