

Transformed material coefficients for single-domain $0.67\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-0.33\text{PbTiO}_3$ single crystals under differently defined coordinate systems

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Due to the confusion in choosing coordinate systems, there occurred some misrepresentations/calculations of effective material properties of $0.67\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-0.33\text{PbTiO}_3$ single crystals based on its single-domain data. This letter is to clarify these confusions by providing proper data set for each of the currently used coordinate systems. © 2004 American Institute of Physics. [DOI: 10.1063/1.1842365]

In order to understand the physical origin of the super-high electromechanical properties found in the multidomain $[001]$ poled $x\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-(1-x)\text{PbTiO}_3$ [PMN-PT] and $x\text{Pb}(\text{Zn}_{1/3}\text{Nb}_{2/3})\text{O}_3-(1-x)\text{PbTiO}_3$ [PZN-PT] single crystals,¹ it is important to know the single-domain data in the rhombohedral phase. Using such single-domain data, one can perform orientation transformation of the property matrix to find out if the origin of the super-high piezoelectric properties is from the orientation effects. For application purposes, if the single-domain data are known, one can calculate the optimum orientational cut to maximize the desired electromechanical properties. For this purpose, a complete set of material coefficients of single-domain $0.67\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-0.33\text{PbTiO}_3$ (PMN-0.33PT) crystals in the rhombohedral $3m$ phase has been measured recently.² Based on this complete set of single-domain properties, it is easy to calculate the rotated material properties and to reach the conclusion that more than 80% of the superior electromechanical properties of the PMN-PT come from orientation effect, i.e., from crystal anisotropy and the large shear piezoelectric coefficient in the single-domain state.^{3,4} We have got many inquiries about these results because some researchers have encountered difficulties trying to duplicate these calculations. One of the causes is the coordinate mislabeling in the original publications³⁻⁵ and the other is the uncertainty of making the one-to-one correspondence between the intrinsic rhombohedral coordinates and the prototype cubic coordinates. The cubic coordinates labeling scheme has been widely used by crystal growers, device designers, and most of the researchers in this field.

While finding the correspondence between the tetragonal phase intrinsic coordinate systems and the cubic coordinate system may not be a problem [the a , $b(=a)$ and c axes in the tetragonal phase can be easily matched to $[100]_c$, $[010]_c$, and $[001]_c$ of the cubic coordinates with the polarization direction along $[001]_c$], such correspondence can be confusing for the rhombohedral ferroelectric phase since there are several different choices of the intrinsic rhombohedral coordinates. Accurately speaking, the angles between any two axes in the rhombohedral unit cell are not 90° , so that the coordinate correspondence is meaningful only between material coordinates (coordinates fixed with the materials and deform with

it). The data reported in Ref. 2 were based on a particular choice of coordinates and the correspondence to cubic coordinates was not properly labeled. Hence, many inquiries and discussions had taken place regarding these rotated values. In order to avoid further confusion, we provide here a table for the complete sets of materials properties in different coordinate systems. This letter provides one-to-one coordinate correspondence between each rhombohedral coordinate system and the cubic coordinate system and also provides a database for interested researchers and engineers.

Shown in Fig. 1 is the recently adopted notation to link the cubic coordinates with the rhombohedral coordinates.⁶ For convenience, we name it the “standard coordinates”. The directions are labeled with a subscript c or r to represent that the directions are referred to cubic or rhombohedral coordinate systems, respectively. The data reported in Ref. 2 were based on a coordinate system rotated 180° about the $[111]_c$ from the standard coordinate system. Two other right-hand coordinate systems are those rotated 90° and 270° about the $[111]_c$, respectively, from the “standard system” shown in Fig. 1. Material properties in different coordinate systems can be transformed by matrix rotation from the standard set given in Table I for PMN-0.33PT single-domain single crystals. Those materials properties that will change signs or values in the other three coordinate systems are listed in Table II for clearness. Properties not listed in Table II are independent of coordinate selection. The four coordinate systems are labeled as 0° , 90° , 180° , and 270° rotated about the polarization direction of $[111]_c$ with the 0° rotated coordinates being the standard coordinates. The explicit correspondence between the possible four choices of rhombohedral coordinate

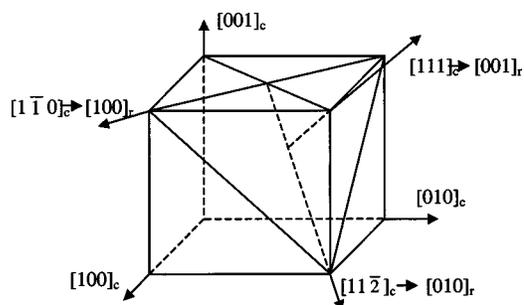


FIG. 1. The standard rhombohedral coordinates defined for PMN-0.33PT and their relationship to the cubic coordinates.

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TABLE I. Measured and derived material properties of single-domain PMN-33%PT single crystal in standard coordinate system.

Elastic stiffness constants: $c_{ij}^E, c_{ij}^D(10^{10} \text{ N/m}^2)$																			
c_{11}^E	c_{12}^E	c_{13}^E	c_{14}^E	c_{15}^E	c_{33}^E	c_{44}^E	c_{66}^E	c_{11}^D	c_{12}^D	c_{13}^D	c_{14}^D	c_{15}^D	c_{33}^D	c_{44}^D	c_{66}^D				
20.12	7.36	11.5	-4.15	0.0	17.12	2.90	6.38	20.43	7.62	10.32	-3.87	0.0	21.94	5.63	6.41				
Elastic compliance constants: $s_{ij}^E, s_{ij}^D(10^{-12} \text{ m}^2/\text{N})$																			
s_{11}^E	s_{12}^E	s_{13}^E	s_{14}^E	s_{15}^E	s_{33}^E	s_{44}^E	s_{66}^E	s_{11}^D	s_{12}^D	s_{13}^D	s_{14}^D	s_{15}^D	s_{33}^D	s_{44}^D	s_{66}^D				
62.16	-53.85	-5.58	166.24	0.0	13.34	510.98	232.02	9.39	-3.94	-2.57	9.15	0.0	6.97	30.33	26.65				
Piezoelectric constants: $e_{i\lambda}(C/\text{m}^2)d_{i\lambda}(10^{-12} \text{ C/N})g_{i\lambda}(10^{-2}\text{Vm/N})h_{i\lambda}(10^8 \text{ V/m})$																			
e_{11}	e_{15}	e_{22}	e_{31}	e_{33}	d_{11}	d_{15}	d_{22}	d_{31}	d_{33}	g_{11}	g_{15}	g_{22}	g_{31}	g_{33}	h_{11}	h_{15}	h_{22}	h_{31}	h_{33}
0.0	7.52	-0.78	-2.88	11.83	0.0	4100	-1340	-90	190	0.0	11.72	-3.83	-1.59	3.35	0.0	36.38	-3.78	-9.94	40.78
Dielectric constants: $\epsilon(\epsilon_0)\beta(10^{-4}/\epsilon_0)$							Electromechanical coupling constants												
ϵ_{11}^S	ϵ_{33}^S	ϵ_{11}^T	ϵ_{33}^T	β_{11}^S	β_{33}^S		β_{11}^T	β_{33}^T	k_{15}	k_{31}	k_{33}	k_t	$\rho(\text{kg/m}^3)$						
233	328	3950	640	42.84	30.53		2.53	15.63	0.70	0.15	0.69	0.47	8060						

systems with its cubic counterpart is given as follows.

(1) Standard coordinates, or 0° rotated coordinates:

$$[100]_r \leftrightarrow [1\bar{1}0]_c, [010]_r \leftrightarrow [11\bar{2}]_c, [001]_r \leftrightarrow [111]_c$$

(2) Coordinates rotated 90° about the $[111]_c$:

$$[100]_r \leftrightarrow [11\bar{2}]_c, [010]_r \leftrightarrow [\bar{1}10]_c, [001]_r \leftrightarrow [111]_c$$

(3) Coordinates rotated 180° about the $[111]_c$:

$$[100]_r \leftrightarrow [\bar{1}10]_c, [010]_r \leftrightarrow [\bar{1}\bar{1}2]_c, [001]_r \leftrightarrow [111]_c$$

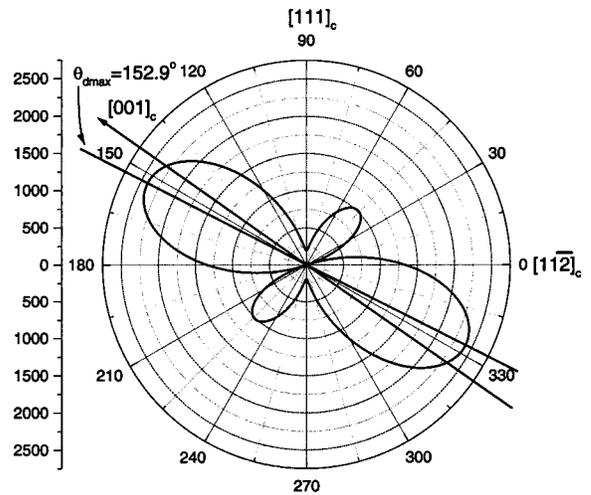
(4) Coordinates rotated 270° about the $[111]_c$:

$$[100]_r \leftrightarrow [\bar{1}\bar{1}2]_c, [010]_r \leftrightarrow [1\bar{1}0]_c, [001]_r \leftrightarrow [111]_c$$

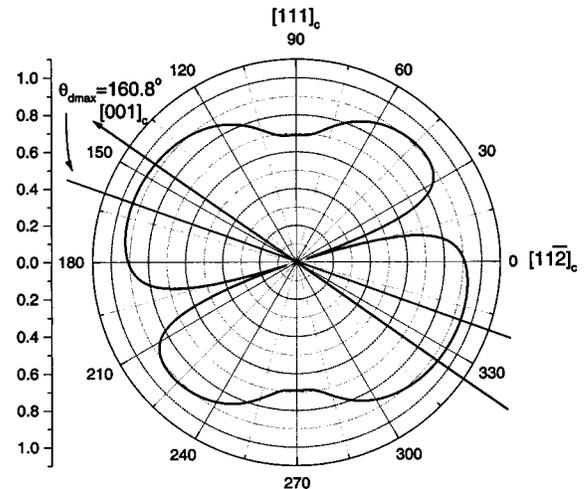
After these coordinate correspondences are defined, it is easy to calculate the optimum cut for the single-domain PMN-33%PT crystals. As we have shown in Ref. 2 that the maximum piezoelectric constant $d_{33}^* = 2411 \text{ pC/N}$ and elec-

TABLE II. Material properties that are changed in the four coordinate systems.

Rotation from standard	Elastic stiffness constants: $c_{ij}^E, c_{ij}^D(10^{10} \text{ N/m}^2)$				Elastic compliance constants: $s_{ij}^E, s_{ij}^D(10^{-12} \text{ m}^2/\text{N})$			
	c_{14}^E	c_{15}^E	c_{14}^D	c_{15}^D	s_{14}^E	s_{15}^E	s_{14}^D	s_{15}^D
0°	-4.15	0.0	-3.87	0.0	166.24	0.0	9.15	0.0
90°	0.0	4.15	0.0	3.87	0.0	-166.24	0.0	-9.15
180°	4.15	0.0	3.87	0	-166.24	0.0	-9.15	0.0
270°	0.0	-4.15	0.0	-3.87	0.0	166.24	0.0	9.15
Rotation from standard	Piezoelectric constants: $e_{i\lambda}(C/\text{m}^2)d_{i\lambda}(10^{-12} \text{ C/N})g_{i\lambda}(10^{-2}\text{Vm/N})h_{i\lambda}(10^8 \text{ V/m})$							
	e_{11}	e_{22}	d_{11}	d_{22}	g_{11}	g_{22}	h_{11}	h_{22}
0°	0.0	-0.78	0.0	-1340	0.0	-3.83	0.0	-3.78
90°	-0.78	0.0	-1340	0.0	-3.83	0.0	-3.78	0.0
180°	0.0	0.78	0.0	1340	0.0	3.83	0.0	3.78
270°	0.78	0.0	1340	0.0	3.83	0.0	3.78	0.0



(a)



(b)

FIG. 2. Cross section plot of orientation dependence of (a) piezoelectric constant d_{33} and (b) electromechanical coupling factor k_{33} of PMN-0.33PT calculated based on single-domain data in Table I.

tromechanical coupling factor $k_{33}^*=0.94$ occur, respectively, in directions 62.9° and 70.8° canted from the spontaneous polarization direction $[111]_c$. The direction of poling for the multidomain crystals, or $[001]_c$, is 54.73° canted from $[111]_c$. Because the d_{33} and k_{33} values are only weakly dependent on orientation near their optimum direction, the calculated effective constants $d_{33}=2316$ pC/N and $k_{33}=0.93$, respectively, in $[001]_c$ are also very large. In fact, it is comparable to the measured multidomain properties.⁷ Hence, we conclude that the superior electromechanical properties in $[001]_c$ poled PMN–0.33PT multidomain crystals mainly come from the orientation effect. The super large d_{15} of the single-domain property is the main contributor to the effective d_{33} of the $[001]_c$ poled multidomain crystals.

Based on the data in the standard coordinate system (0° rotated coordinates), the orientation dependence of electromechanical properties has been calculated for PMN–0.33PT single crystals and given in Fig. 2, which is a cross section plot looking down at the $[100]_r$ (or $[1\bar{1}0]_c$). The rotations of piezoelectric constant d_{33} and electromechanical coupling factor k_{33} were performed based on the transformation matrices given by Cady.⁸ The calculated results showed that the

material properties of PMN–0.33PT single-domain crystal are highly anisotropic and the maximum value of effective d_{33} occurs in the directions 152.9° and 332.9° , respectively, from the $[010]_r$ (or $[11\bar{2}]_c$) while the maximum value of k_{33} occurs in directions 160.8° and 340.8° , respectively, from the $[010]_r$. Note here, curves in Fig. 2 are rotated in comparison to what was reported in Ref. 2 due to different choice of coordinates. However, the conclusion obtained in Ref. 2, i.e., the super large effective d_{33} comes from the orientation effect, remains valid.

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