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Inconsistencies of the complete sets of electromechanical constants of relaxor-ferroelectric single crystals

V. Yu. Topolov^{1,a)} and C. R. Bowen²

¹Department of Physics, Southern Federal University, 5 Zorge Street, 344090 Rostov-on-Don, Russia

²Materials Research Centre, Department of Mechanical Engineering, University of Bath, BA2 7AY Bath, United Kingdom

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Complete sets of elastic, dielectric, and piezoelectric constants of relaxor-ferroelectric single crystals of $(1 - x)\text{Pb}(\text{A}_{1/3}\text{Nb}_{2/3})\cdot\text{O}_3 - x\text{PbTiO}_3$ ($\text{A} = \text{Mg}$ or Zn) and $\text{Pb}(\text{In}_{1/2}\text{Nb}_{1/2})\text{O}_3 - \text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\cdot\text{O}_3 - \text{PbTiO}_3$ are analyzed to demonstrate violations of the interrelations between particular groups of electromechanical constants and the resulting influence of the inconsistent properties on the thermodynamic stability and electromechanical coupling in single crystals. The possibilities of refinement and correction of inconsistent constants are discussed and presented for specific examples of relaxor-ferroelectric single crystals. © 2011 American Institute of Physics. [doi:10.1063/1.3581117]

I. INTRODUCTION

Single crystals (SCs) of relaxor-ferroelectric solid solutions¹ with the perovskite-type structure and compositions near the morphotropic phase boundary exhibit outstanding piezoelectric performance and electromechanical coupling in comparison with conventional ferroelectric ceramics. The ferroelectric-relaxor SCs, such as $(1 - x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\cdot\text{O}_3 - x\text{PbTiO}_3$ (PMN- x PT) and $(1 - x)\text{Pb}(\text{Zn}_{1/3}\text{Nb}_{2/3})\cdot\text{O}_3 - x\text{PbTiO}_3$ (PZN- x PT), are of interest for piezoelectric transducer, actuator, sensor, hydrophone, and solid-state electronic applications² as well as piezo-active components of advanced composites.³ For these applications and for modeling SC transducers, it is important to have reliable information on the physical properties of the aforementioned and related SCs. Complete sets of elastic, dielectric, and piezoelectric (i.e., electromechanical) constants are usually determined⁴⁻⁶ for specific SC sample geometries and using the recognized experimental methods. Among the complete sets of constants, of particular interest for many applications are the elastic compliances s_{ab}^E at electric field $E = \text{const}$, piezoelectric coefficients d_{ij} , and dielectric permittivities ϵ_{pp}^T at mechanical stress $T = \text{const}$. Other important complete sets are the elastic moduli c_{ab}^E at electric field $E = \text{const}$, piezoelectric coefficients e_{ij} , and dielectric permittivities ϵ_{mm}^S at mechanical strain $S = \text{const}$. These electromechanical constants from the complete sets are related via the following constitutive equations⁷ of an electroelastic medium:

$$S_p = s_{pq}^E T_q + d_{fp} E_f \quad \text{and} \quad D_k = d_{kl} T_l + \epsilon_{kr}^T E_r \quad (1)$$

or

$$T_p = c_{pq}^E S_q - e_{fp} E_f \quad \text{and} \quad D_k = e_{kl} S_l + \epsilon_{kr}^S E_r, \quad (2)$$

where D_k is electric displacement. There are also two pairs of the constitutive equations⁷ for the additional piezoelectric coefficients, g_{ij} and h_{ij} :

$$s_p = s_{pq}^D T_q + g_{fp} D_f \quad \text{and} \quad E_k = -g_{kl} T_l + \beta_{kr}^T D_r, \quad (3)$$

and

$$T_p = c_{pq}^D S_q - h_{fp} D_f \quad \text{and} \quad E_k = -h_{kl} S_l + \beta_{kr}^S D_r, \quad (4)$$

where β_{kr}^T is dielectric impermeability at $T = \text{const}$ and β_{kr}^S is dielectric impermeability at $S = \text{const}$. The piezoelectric coefficients from Eqs. (1)–(4) are interrelated as follows:

$$d_{fp} = \epsilon_{fk}^T g_{kp} = e_{fq} s_{qp}^E, \quad e_{fp} = \epsilon_{fk}^S h_{kp} = d_{fq} c_{qp}^E, \quad g_{fp} = \beta_{fk}^T d_{kp} \\ = h_{fq} s_{qp}^D, \quad \text{and} \quad h_{fp} = \beta_{fk}^S e_{kp} = g_{fq} c_{qp}^D. \quad (5)$$

The complete sets of room-temperature electromechanical constants of PMN- x PT, PZN- x PT, and $\text{Pb}(\text{In}_{1/2}\text{Nb}_{1/2})\cdot\text{O}_3 - \text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\cdot\text{O}_3 - \text{PbTiO}_3$ (PIN-PMN-PT) SCs have been experimentally determined in both the single-domain and polydomain states and reported in a series of papers (see, e.g., Refs. 4–6). These complete sets refer to fixed poling directions, often to [001], [110] or [111] of the perovskite unit cell. Good agreement, or *good consistency*, between the different sets of electromechanical constants related to the same SC [see Eqs. (1)–(5)] is observed for data on the [001]-poled samples of PMN-0.30PT (Ref. 8) and PIN-PMN-PT (Ref. 9). In these cases, any deviations of constants from Eq. (5) are less than 5%.¹⁰ However, in some cases we have observed issues concerned with an inconsistency¹¹ of the electromechanical constants and related parameters of SCs. In the context of this paper, an ‘inconsistency’ is expressed as a 5% or greater disagreement between the values of electromechanical constants from the related complete sets. Examples of inconsistency include the transition from elastic compliances s_{ab}^E to elastic moduli c_{ab}^E , from the piezoelectric coefficients e_{kp} to the piezoelectric coefficients h_{fp} [see Eq. (5)], etc. An inconsistency can also represent a violation of conditions for the thermodynamic stability⁷ of SCs.

The aim of this paper is to demonstrate examples of the inconsistencies for relaxor-ferroelectric SCs and to discuss

^{a)}Electronic mail: vutopolov@sfnu.ru.

the possibilities of the refinement of particular electromechanical constants of these materials.

II. EXAMPLES OF INCONSISTENCY AND REFINEMENT

A. PZN-xPT

The complete sets of electromechanical constants of PZN-(0.06–0.07)PT SCs in the single-domain¹² and polydomain¹³ states are characterized by a particular inconsistency. In case of the single-domain SC, this inconsistency is concerned with interrelations between different electromechanical constants, mainly, between the piezoelectric coefficients from Eq. (5). Below we consider some quantitative interrelations between the constants listed in Table I.

Taking into account the link between the piezoelectric coefficients h_{fp} and e_{kp} from Eq. (5), the equality $\|\beta^S\| = \|\epsilon^S\|^{-1}$, and the $3m$ symmetry of the single-domain PZN-(0.06–0.07)PT SC¹² at room temperature, we obtain

$$h_{22} = e_{22}/\epsilon_{22}^S > 0. \quad (6)$$

However, Table I contains $h_{22} < 0$ and $e_{22} > 0$. Such a discrepancy can stem from an erroneous sign of dielectric permittivity ϵ_{22}^S at the frequency of measurements,¹² but our evaluation suggests that h_{22} from Eq. (6) would be 8.3 times more than $|h_{22}|$ from Table I. Using the relation (Ref. 7) $h_{33} = e_{33}/\epsilon_{33}^S$, we obtain $h_{33} = 25.1 \cdot 10^8$ V/m, and the difference between this value and h_{33} from Table I is about 8.7%. The evaluation of h_{31} and h_{15} from formulas (Ref. 7) $h_{31} = e_{31}/\epsilon_{33}^S$ and $h_{15} = e_{15}/\epsilon_{11}^S$ leads to $h_{31} = -9.17 \cdot 10^8$ V/m and $h_{15} = 8.48 \cdot 10^8$ V/m. Both of these values are in disagreement with the h_{31} and h_{15} values from Table I, where $h_{31} = -5.9$ and $h_{15} = 19.7$ (in 10^8 V/m).

We have also checked quantitative relations between the piezoelectric coefficients h_{fp} and g_{fq} from Eq. (5). Good agreement between the evaluated and published¹² values of h_{33} ($22.6 \cdot 10^8$ V/m from our evaluation) is observed, but there is a significant difference between the remaining piezoelectric coefficients h_{fp} . For example, based on our evaluation of g_{fq} and c_{qp}^D , the piezoelectric coefficients $h_{31} = -6.84 \cdot 10^8$ V/m (difference about 16%) and $h_{15} = 30.9 \cdot 10^8$ V/m (difference about 57%) indicate the presence of the inconsistency in the data of Table I.

On checking the link between the piezoelectric coefficients e_{fp} and d_{fq} from Eq. (5), there is good agreement between the evaluated and published¹² values of e_{3p} ($p = 1$ and 3); while the difference in the e_{15} values ($e_{15} = 29.4$ C/m² from our evaluation) is 7.8%. From the link between the piezoelectric coefficients g_{fp} and d_{kp} [see Eq. (5)], we obtain $g_{22} = d_{22}/\epsilon_{22}^T = -1.32 \cdot 10^{-2}$ V · m/N < 0 , but according to Table I, $g_{22} = 1.3 \cdot 10^{-2}$ V · m/N. The remaining values of g_{fp} , evaluated using d_{kp} , are in agreement with data shown in Table I.

Next, we consider some interrelations that involve electromechanical constants related to the elastic, piezoelectric, and dielectric properties of SCs. As is known for a piezoelectric medium,⁷ its elastic moduli c_{33}^D and c_{33}^E are linked by an equality $c_{33}^D - c_{33}^E = e_{33}h_{33}$. Taking the values of these constants from Table I, we observe a difference between $c_{33}^D - c_{33}^E$ and $e_{33}h_{33}$ as large as 6.9%; while the thickness electromechanical coupling factor $k_t = [(c_{33}^D - c_{33}^E)/c_{33}^D]^{1/2}$ of 0.392 is in good agreement with the k_t value from Table I. Additionally, k_t evaluated using the conventional formula⁷

$$k_t = e_{33}/(\epsilon_{33}^S c_{33}^D)^{1/2} \quad (7)$$

equals 0.396 and is consistent with data from Table I.

Of independent interest are evaluations of electromechanical coupling factors⁷

$$k_{ij} = d_{ij}/(\epsilon_{ii}^T \epsilon_{jj}^E)^{1/2}. \quad (8)$$

Taking the pertinent electromechanical constants from Table I and using Eq. (8), we obtain $k_{33} = 0.46$, $|k_{31}| = 0.078$, and $k_{15} = 0.97$, which are different to the values in Table I, where $k_{33} = 0.33$, $|k_{31}| = 0.18$, and $k_{15} = 0.73$. This and the above-given examples of our evaluations suggest that the inconsistency of electromechanical constants¹² of the single-domain PZN-(0.06–0.07)PT SC cannot be avoided at a re-measurement of several constants, and the complete set of the electromechanical properties is needed to be carefully refined on high-quality samples in a relatively stable single-domain state.

Analyzing the complete set of electromechanical constants of the polydomain PZN-(0.06–0.07)PT SCs,¹³ we observe an inconsistency at the transition from elastic compliances s_{qp}^E and piezoelectric coefficients d_{fp} to e_{fq} [see

TABLE I. Elastic compliances s_{ab}^E (in 10^{-12} Pa⁻¹), elastic moduli c_{ab}^E and c_{ab}^D (in 10^{10} Pa), piezoelectric coefficients d_{ij} (in pC/N), e_{ij} (in C/m²), g_{ij} (in 10^{-2} V · m/N), and h_{ij} (in 10^8 V/m), relative dielectric permittivities $\epsilon_{pp}^S/\epsilon_0$ and $\epsilon_{pp}^T/\epsilon_0$, and electromechanical coupling factors k_{ij} and k_t of single-domain PZN-(0.06–0.07)PT SC (Ref. 12).

s_{11}^E	s_{12}^E	s_{13}^E	s_{14}^E	s_{33}^E	s_{44}^E	s_{66}^E	c_{11}^E	c_{12}^E
32.8	-28.5	-1.2	100.1	6.5	390.4	122.6	18.0	8.0
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
4.8	-2.6	17.1	1.6	5.0	19.4	8.0	6.4	-2.2
c_{33}^D	c_{44}^D	c_{66}^D	d_{15}	d_{22}	d_{31}	d_{33}	e_{15}	e_{22}
20.2	4.0	5.7	6000	-1280	-35	93	31.7	30.9
e_{31}	e_{33}	g_{15}	g_{22}	g_{31}	g_{33}	h_{15}	h_{22}	h_{31}
-4.6	12.6	6.3	1.3	-0.6	1.5	19.7	-1.0	-5.9
h_{33}	$\epsilon_{11}^S/\epsilon_0$	$\epsilon_{33}^S/\epsilon_0$	$\epsilon_{11}^T/\epsilon_0$	$\epsilon_{33}^T/\epsilon_0$	k_{15}	k_{31}	k_{33}	k_t
23.1	4222	567	11000	700	0.73	0.18	0.33	0.39

Eq. (5)]. According to our evaluations, $e_{31} < 0$, $e_{33} < 0$, and $|e_{31}| > |e_{33}|$, i.e., these constants are in obvious disagreement with those from Ref. 13.

Recent work¹⁴ on the performance of the $[110]^L \times [001]^T$ cuts of the polydomain relaxor-ferroelectric SCs poled along [001] of the perovskite unit cell contains some complete sets of electromechanical constants deduced using the method put forward by Shukla *et al.* Analyzing these constants, one can observe the inconsistency at the transition from the elastic compliances s_{ab}^E to elastic moduli c_{ab}^E and piezoelectric coefficients e_{fq} . The interrelation between elastic constants⁷ is represented in the matrix form as

$$\|c^E\| = \|s^E\|^{-1}, \quad (9)$$

and the piezoelectric coefficients e_{fq} are evaluated in accordance with Eq. (5). Data from Table II show that both the elastic and piezoelectric constants of PZN-0.045PT SC¹⁴ are characterized by an inconsistency ($\delta > 5\%$ in Table III). This can be a result of the influence of the domain structure and defects in SCs on the electromechanical properties, especially as they are measured in nonpolar directions and on specific SC cuts.

B. PIN-PMN-PT

The characteristics of the piezoelectric and dielectric properties of single-domain [001]-poled PIN-PMN-PT SCs near the morphotropic phase boundary, namely the high electromechanical coupling and the stability of the single-domain state⁶ may be useful for future piezotechnical and smart-materials applications. However, a comparison of data⁶ on the tetragonal single-domain PIN-PMN-PT SC (measured and calculated properties) suggests inconsistencies among the electromechanical constants of the studied composition. Examples of the inconsistency (Table III) concern different kinds of electromechanical constants. Values of X (see the second column in Table III) were calculated using Eq. (5) and taking into account tetragonal symmetry of

TABLE II. Elastic moduli c_{ab}^E (in 10^{10} Pa)^a and piezoelectric coefficients e_{ij} (in C/m^2)^b of [001]-poled $[110]^L \times [001]^T$ cuts of polydomain PZN-0.045PT SC.

Electromechanical constant X to be evaluated using data from Ref. 14	Value of X (current evaluation)	Value of X_{tab} from Ref. 14	$\delta = X - X_{tab} / X_{tab}$, %
c_{11}^E	15.7	17.0	6.4
c_{12}^E	3.05	4.35	30
c_{13}^E	8.85	10.1	12
c_{33}^E	9.29	10.5	12
c_{44}^E	6.41	6.4	< 1
c_{66}^E	0.452	0.45	< 1
e_{31}	-4.79	-3.7	< 1
e_{33}	14.0	15.0	6.3
e_{15}	8.97	8.9	< 1

^aEvaluated using Eq. (9).

^bEvaluated using Eq. (5).

TABLE III. Published (X_{tab}) and refined (X) values of electromechanical constants determined for [001]-poled PIN-PMN-PT SCs.

Electromechanical constant X to be evaluated using data from Ref. 6 and formulas from Ref. 7	Value of X (current evaluation)	Value of X_{tab} from Ref. 6	$\delta = X - X_{tab} / X_{tab}$, %
c_{11}^E (in 10^{10} Pa) using s_{ab}^E	15.1	20.6	27
c_{12}^E (in 10^{10} Pa) using s_{ab}^E	10.2	15.5	34
c_{13}^E (in 10^{10} Pa) using s_{ab}^E	8.7	12.5	30
c_{33}^E (in 10^{10} Pa) using s_{ab}^E	8.5	12.5	32
c_{44}^E (in 10^{10} Pa) using s_{ab}^E	1.8	1.8	0
c_{66}^E (in 10^{10} Pa) using s_{ab}^E	4.0	4.5	11
c_{11}^D (in 10^{10} Pa) using s_{ab}^D	14.6	21.3	31
c_{12}^D (in 10^{10} Pa) using s_{ab}^D	9.4	16.3	42
c_{13}^D (in 10^{10} Pa) using s_{ab}^D	5.0	10.9	54
c_{33}^D (in 10^{10} Pa) using s_{ab}^D	10.3	19.4	47
c_{44}^D (in 10^{10} Pa) using s_{ab}^D	6.6	6.8	2.9
c_{66}^D (in 10^{10} Pa) using s_{ab}^D	4.0	4.5	11
e_{31} (in C/m^2) using s_{ab}^E and d_{ij}	-4.1	-4.2	2.4
e_{33} (in C/m^2) using s_{ab}^E and d_{ij}	10.1	9.5	6.3
e_{15} (in C/m^2) using s_{ab}^E and d_{ij}	42.7	46.4	8.0
$\epsilon_{11}^S / \epsilon_0$ using e_{pp}^T , s_{ab}^E , and d_{ij}	3650	4800	24
$\epsilon_{33}^S / \epsilon_0$ using e_{pp}^T , s_{ab}^E , and d_{ij}	300	310	3.2
h_{31} (in 10^8 N/C) using c_{ab}^D and g_{ij}	-17.5	-16.5	6.1
h_{33} (in 10^8 N/C) using c_{ab}^D and g_{ij}	61.8	34.9	77
h_{15} (in 10^8 N/C) using c_{ab}^D and g_{ij}	11.2	10.8	3.7
h_{31} (in 10^8 N/C) using e_{ij} and e_{pp}^S	-15.3	-16.5	7.3
h_{33} (in 10^8 N/C) using e_{ij} and e_{pp}^S	34.6	34.9	0.9
h_{15} (in 10^8 N/C) using e_{ij} and e_{pp}^S	10.9	10.8	0.9

the studied PIN-PMN-PT composition. It is important to emphasize the large difference between the calculated values (X) of elastic constants and those reported⁶ (see X_{tab} in Table III). This concerns, for example, the elastic moduli c_{ab}^E and c_{ab}^D from the fourth column in Table III. Elastic constants involve expressions⁷ suitable for the determination of dielectric and piezoelectric properties, and the influence of inconsistent elastic properties on these properties is also observed in Table III [see, e.g., data on $\epsilon_{11}^S/\epsilon_0$ and h_{33} calculated using c_{qp}^D and g_{fq} in accordance with Eq. (5)]. At the same time, the low differences between values of X and X_{tab} (less than 1% in Table III) suggest that the procedure⁶ of the determination of electromechanical constants allows one to avoid any inconsistency by careful consideration of measurement data on the studied PIN-PMN-PT SCs.

C. PMN-xPT

Very recently, examples of the inconsistency among electromechanical constants were analyzed¹¹ for polydomain [011]-poled SCs of PMN-xPT¹⁵ with $x = 0.28, 0.30$, and 0.32 . The complete set of electromechanical constants was also measured¹⁶ on [001]-poled domain-engineered PMN-0.28PT samples with macroscopic tetragonal ($4mm$) symmetry at room temperature. Elastic constants c_{ab}^E and s_{ab}^E from Ref. 16 are consistent as stated using Eq. (9). However, the piezoelectric coefficients e_{3j} calculated using Eq. (5) and values of c_{ab}^E and d_{ij} from paper¹⁶ differ considerably from the piezoelectric coefficients e_{ij} published in the same paper.¹⁶

For instance, conditions $e_{33} < 0$ and $|e_{33}| < |e_{31}|$ are achieved in our calculations based on Eq. (5), while the piezoelectric coefficients¹⁶ obey inequalities $e_{33} > 0$ and $e_{33} > |e_{31}|$. Electromechanical constants from Ref. 16 are also inconsistent because of an overestimated value of the longitudinal electromechanical coupling factor k_{33} from Eq. (8). Taking $d_{33} = 2365$ pC/N, $\epsilon_{33}^T/\epsilon_0 = 6833$, and $s_{33}^E = 86.46 \cdot 10^{-12}$ Pa⁻¹ from paper¹⁶ and substituting these values into right part of Eq. (8), we obtain $k_{33} = 2365/2287 > 1$, that clearly does not have any physical justification. This highlights the potential future problems of using the complete sets of electromechanical constants¹⁶ of the polydomain PMN-0.28PT SC.

An important example of the inconsistency is revealed after a careful analysis of experimental data⁴ on [001]-poled PMN-0.38PT SCs. According to results,⁴ this SC has tetragonal symmetry at room temperature, however, no specifics on domain structure (single-domain, near single-domain, polydomain, etc.) is provided. Our comments on the electromechanical constants from Ref. 4 are given as follows. First, elastic moduli of PMN-0.38PT SC (see c_{11}^E , c_{12}^E , and c_{13}^E from Table IV and c_{33}^E from footnote b after Table IV) do not obey the condition for the thermodynamic stability¹⁷

$$(c_{11}^E + c_{12}^E)c_{33}^E > 2(c_{13}^E)^2. \quad (10)$$

Second, a transition from $\|s^E\|$ to $\|c^E\|$ in accordance with Eq. (9) enables us to obtain $c_{33}^E = 14.89 \cdot 10^{10}$ Pa instead of $c_{33}^E = 9.92 \cdot 10^{10}$ Pa. The set of refined elastic moduli c_{ab}^E from Table IV obeys inequality, Eq. (10), and other related conditions^{7,17} for stability. Based on this set of refined data, one can confirm full agreement between the piezoelectric coefficients e_{3j} calculated using Eq. (5) and those determined in Ref. 4. Correctness of values⁴ of the piezoelectric coefficients d_{3j} and e_{3j} is also confirmed when checking a link between dielectric permittivities⁷

$$\epsilon_{pp}^T - \epsilon_{pp}^S = d_{pj}e_{pj}. \quad (11)$$

It is also observed that a value of $d_{15}e_{15}$ from Eq. (11) at $p = 1$ does not correspond to the difference $\epsilon_{11}^T - \epsilon_{11}^S$ that is calculated from data published in paper of Ref. 4.

Third, our calculation of the electromechanical coupling factor k_{33} [see Eq. (8)] using d_{33} , ϵ_{33}^T , and s_{33}^E from experimental data⁴ leads to $k_{33} = 0.802$ that is less than k_{33} from Ref. 4. The value of the thickness electromechanical coupling factor k_t from Eq. (7) is evaluated taking into account

TABLE IV. Refined electromechanical constants of [001]-poled PMN-0.38PT SC. Elastic moduli c_{ab}^E and c_{33}^D are in 10^{10} Pa.

c_{11}^E	c_{12}^E	c_{13}^E	c_{33}^E	c_{44}^E	c_{66}^E	c_{33}^D
21.25 ^a	14.33 ^a	13.51 ^a	14.89 ^b	5.56 ^a	6.95 ^a	20.72 ^c
$\epsilon_{11}^S/\epsilon_0$	$\epsilon_{33}^S/\epsilon_0$	$\epsilon_{11}^T/\epsilon_0$	$\epsilon_{33}^T/\epsilon_0$	$ k_{31} $	k_{33}	k_t
3395 ^d	255 ^a	4301 ^a	734 ^a	0.446 ^a	0.802 ^c	0.530 ^f

^aData from Ref. 4.

^bInstead of $c_{33}^E = 9.92 \cdot 10^{10}$ Pa from Ref. 4.

^cInstead of $c_{33}^D = 15.74 \cdot 10^{12}$ Pa from Ref. 4.

^dInstead of $\epsilon_{11}^S/\epsilon_0 = 2171$ from Ref. 4.

^eInstead of $k_{33} = 0.846$ from Ref. 4.

^fInstead of $k_t = 0.608$ from Ref. 4.

e_{33} , ϵ_{33}^S from Eq. (11), and a relation⁷ $c_{33}^D = c_{33}^E + e_{33}h_{33}$. This k_t value becomes 14.7% less than the k_t value from paper⁴ (see Table IV). The main reason for such a difference is associated with the refined (larger) elastic modulus c_{33}^E and its influence on c_{33}^D and, therefore, on k_t from Eq. (7).

Electromechanical constants were also found¹⁴ for the $[110]^L \times [001]^T$ cut of the polydomain PMN-0.28PT SC poled along [001]. It seems probable that elastic compliance s_{13}^E equals $-37.8 \cdot 10^{-12}$ Pa⁻¹ instead of $37.8 \cdot 10^{-12}$ Pa⁻¹ from Ref. 14. Our subsequent checking the interrelations between elastic and piezoelectric constants enables us to conclude that the largest differences between the elastic moduli evaluated using Eq. (9) and those from paper (see Ref. 14) are attained for c_{11}^E (about 12%) and c_{33}^E (about 7.4%). Along with these differences, the considerable discrepancy is revealed when comparing the values of the piezoelectric coefficients e_{3j} . Our evaluation based on Eq. (5) would lead to $e_{31} = -17.7$ and $e_{33} = 8.22$ (in C/m²), while the published¹⁴ values are $e_{31} = -4.7$ and $e_{33} = 20.5$ (in C/m²). In contrast to this, a difference between the e_{15} values determined in the same way is only about 0.2%. Such discrepancies may stimulate careful experimental determination of the elastic and piezoelectric properties of the polydomain PMN-0.28PT SC¹⁴ at various orientations of its main crystallographic axes.

III. CONCLUSIONS

This paper reports a few examples of the inconsistency in the complete sets of room-temperature electromechanical constants of advanced relaxor-ferroelectric SCs. The inconsistent constants violate Eqs. (5), (7)–(9), and (11) for the piezoelectric medium, and Eq. (10) for thermodynamic stability of SCs. Moreover, the inconsistency can lead to an overestimation of the electromechanical coupling factors. The inconsistency reported in this paper is accounted for by different reasons (measurement conditions, accuracy of measurement, quantitative treatment of measured data,^{10,11} etc.) and can be corrected in several cases. Our results of correction (Tables II–IV) suggest that the difference between the inconsistent and consistent values can reach a few tens of a percent which can be significant in transducer design and modeling of transducer behavior. In particular, it concerns the electromechanical constants of the single-domain PZN-(0.06–0.07)PT¹² and PIN-PMN-PT⁶ SCs, and further careful measurements of their properties are desirable. The further refinement of data^{12–14,16} is also needed for PMN-0.28PT SCs. As for PMN-0.38PT SC, the corrected set of electromechanical constants (Table IV) can be suitable for further studies, design purposes and prediction of effective parameters. Of independent interest are conditions for stability of the single-domain state in PIN-PMN-PT, PZN-(0.06–0.07)PT, and PMN-0.38PT SCs. It is believed that this problem may be considered, and inconsistencies will be avoided in experimental studies and piezotechnical applications in the future.

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- ¹L. E. Cross, *Piezoelectricity. Evolution and Future of a Technology*, edited by W. Heywang, W. Wersing, and K. Lubitz (Springer, Berlin, 2008), p. 132; B. Noheda, *Curr. Opin. Solid State Mater. Sci.* **6**, 27 (2002).
- ²K. C. Cheng, H. L. W. Chan, C. L. Choy, Q. Yin, H. Luo, and Z. Yin, *IEEE Trans. Ultrason. Ferroelectr. Freq. Control* **50**, 1177 (2003); K. Ren, Y. Liu, X. Geng, H. F. Hofmann, and Q. M. Zhang, *IEEE Trans. Ultrason. Ferroelectr. Freq. Control* **53**, 631 (2006).
- ³V. Yu. Topolov and C. R. Bowen, *Electromechanical Properties in Composites Based on Ferroelectrics* (Springer, London, 2009).
- ⁴H. Cao and H. Luo, *Ferroelectrics* **274**, 309 (2002).
- ⁵X. Liu, S. Zhang, J. Luo, T. R. Shrout, and W. Cao, *Appl. Phys. Lett.* **96**, 012907 (2010).
- ⁶F. Li, S. Zhang, Z. Xu, X. Wei, J. Luo, and T. R. Shrout, *J. Appl. Phys.* **107**, 054107 (2010).
- ⁷T. Ikeda, *Fundamentals of Piezoelectricity* (Oxford University Press, New York, 1990).
- ⁸R. Zhang, W. Jiang, B. Jiang, and W. Cao, *Fundamental Physics of Ferroelectrics 2002*, edited by R. E. Cohen (American Institute of Physics, Melville, NY, 2002), p. 195.
- ⁹S. Zhang, J. Luo, W. Hackenberger, and T. R. Shrout, *J. Appl. Phys.* **104**, 064106 (2008).
- ¹⁰As follows from the *IEEE Standard on Piezoelectricity 176-1987* (IEEE, New York, NY, 1988), different techniques for measuring the piezoelectric coefficients are capable of reasonable accuracy, a few percent or less under optimum conditions. According to the European Standard EN 50324-2:2002 "*Piezoelectric Properties of Ceramic Materials and Components – Part 2: Methods of Measurement – Low Power*" (CENELEC, Brussels, 2002), the accuracy of the calculation of the complete set of material coefficients depends on material variability and measuring accuracy. In our study we assume that the consistency takes place as electromechanical constants from Eqs. (1)–(4) obey constitutive formulas for the piezoelectric medium (see Ref. 7) with accuracy to 5%.
- ¹¹V. Yu. Topolov, *Appl. Phys. Lett.* **96**, 196101 (2010).
- ¹²J. Jin, K. K. Rajan, and L. C. Lim, *Jap. J. Appl. Phys.* **45**, 8744 (2006).
- ¹³R. Shukla, K. K. Rajan, P. Gandhi, and L.-C. Lim, *Appl. Phys. Lett.* **92**, 212907 (2008).
- ¹⁴R. Shukla, K. K. Rajan, M. Shanthi, J. Jin, L.-C. Lim, and P. Gandhi, *J. Appl. Phys.* **107**, 014102 (2010).
- ¹⁵M. Shanthi, L. C. Lim, K. K. Rajan, and J. Jin, *Appl. Phys. Lett.* **92**, 142906 (2008).
- ¹⁶D. Zhou, F. Wang, L. Luo, J. Chen, W. Ge, X. Zhao, and H. Luo, *J. Phys. D: Appl. Phys.* **41**, 185402 (2008).
- ¹⁷J. Nye, *Physical Properties of Crystals* (Clarendon, Oxford, 1964).