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POLARIZATION MATRICES OF LITHIUM TETRABORATE

ARTHUR BALLATO ELECTRONICS TECHNOLOGY AND DEVICES LABORATORY

JUNE 1989

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INTRODUCTION

Electromechanical transduction taking place via the piezoelectric effect is characterized phenomenologically by constitutive equations that relate the elastic and electric variables. These equations take a variety of forms, depending upon the choice of independent and dependent variables; the choice is normally dictated by the application. For example, piezoelectric resonators in the form of thickness mode plates are most easily treated using the isagric elastic stiffnesses [cE], the piezoelectric stress constants [e], and the dielectric permittivities at constant strain [(eps)S].

Various measurement techniques yield values for the elements of a particular coefficient set more directly than those of another. The coefficients appearing in the different equation sets are, however, interrelated, so that once any one complete set is available, all the other sets of elements may be found. The most accurate and precise experimental results to date have been from plate resonator (resonance) and pulse-echo (transit-time) measurements. From the [cE], [e], and [(eps)S] matrices determined therefrom, those matrices representing material properties expressed in the other alternative forms may be calculated.

Electrooptical applications are becoming increasingly important. So also are treatments of piezoelectric and ferroelectric phenomena from the standpoint of molecular interactions. In both of these cases the constitutive equations using polarization as the independent electrical variable, rather than either electric intensity or displacement, assume greater importance than the sets traditionally used for transducer, signal processing, and resonator applications.

In this report we give the complete sets of linear constitutive equations relating elastic and electric fields. For each equation set the numerical values are computed for lithium tetraborate from the measured [sE], [d], and [(eps)T] values of Shiosaki, et al. (Ref.1). Coupling to the thermal field is neglected. Rationalized mks units are used throughout.

CONSTITUTIVE EQUATION SETS

Symbols and units for the quantities employed are given in Table 1. In terms of these, six constitutive equation sets are used. Of these, electric intensity, dielectric displacement, and polarization each appear in two sets as an independent variable. The sets are, in compressed matrix notation, as follows. A prime denotes transpose; [I] is the unit matrix.

I. The Piezoelectric Stress Constant Set

[T]	=	[CE] [S]	-	[e]'	[E]	(1)
[D]	=	[e][S]	+	[(eps)S]	[E]	(2)

QUANTITY	UNIT	SYMBOL/DEFINITION
Elastic stress	N/m2	[T]
Elastic strain		[S]
Electric intensity	V/m	[E]
Dielectric displacement	C/m2	[D]
Dielectric polarization	C/m2	[P]
Elastic compliance at constant [E], [D], [P]	m2/N	[CE], [CD], [CP]
Elastic stiffness at constant [E], [D], [P]	N/m2	[sE], [sD], [sP]
Dielectric permittivity at constant [T], [S]	F/m	[(eps)T], [(eps)S]
Dielectric constant, relative, at constant [T], [S]		[(Kr)T], [(Kr)S] =[(eps)T]/(eps)o, [(eps)S]/(eps)o
Dielectric impermeability at constant [T], [S]	m/F	[(bet)T], [(bet)S] =[(eps)T] (-1), [(eps)S] (-1)
Dielectric impermeability, relative, at constant [T], [S]		<pre>[(betr)T], [(betr)S] =[(bet)T]*(eps)o, [(bet)S]*(eps)o =[(Kr)T] (-1), [(Kr)S] (-1)'</pre>
Dielectric susceptibility at constant [T], [S]	F/m	[(chi)T], [(chi)S] =[(Kr)T-I]*(eps)o, [(Kr)S-I]*(eps)o
Dielectric susceptibility, relative, at constant [T], [S]		[(chir)T], [(chir)S] =[(chi)T]/(eps)o, [(chi)S]/(eps)o
Reciprocal dielectric susceptibility at constant [T], [S]	m/F	[(zet)T], [(zet)S] =[(chi)T] (-1), [(chi)S] (-1)'
Reciprocal dielectric susceptibility, relative, at constant [T], [S]		[(zetr)T], [(zetr)S] =[(zet)T]*(eps)o, [(zet)S]*(eps)o
Piezoelectric stress	C/m2	[e]
constant	2	

TABLE 1. SYMBOLS, UNITS, AND DEFINITIONS.

TABLE 1. SYMBOL	S, UNITS, A	ND DEFINITIONS.	(continued)
QUANTITY		UNIT	SYMBOL/DEFINITION
Piezoelectric st coefficient	rain	m/V = C/N	[d]
Piezoelectric st modulus	ress	N/C = V/m	[h]
Piezoelectric st constant	rain	m2/C	[d]
Piezoelectric po modulus	larization	V/m = N/C	[a]
Piezoelectric po constant	larization	m2/C	[b]
	===***	=======================================	

Note: Square brackets, <u>sic</u>: [], denote matrices.

II. The Piezoelectric Strain Coefficient Set [d]' (3) [S] [SE] [T] + (E) Ξ [D] = [d] [T]+ [(eps)T] (E) (4) III. The Piezoelectric Stress Modulus Set (5) [T] = [CD] [S] [h]' [D] = -[h][S]+ [(bet)S] (6) [E] [D] The Piezoelectric Strain Constant Set IV. (7) [S] = [sD] [T]+ [g]' [D] [E] = -[q] [T]+ [(bet)T][D] (8) v. The Piezoelectric Polarization Modulus Set (9) **[T]** = [CP] [S]-[a]' [P] = -[a][S]+ [(zet)S](10)(E) [P] VI. The Piezoelectric Polarization Constant Set (11)[S] = [SP] [T] + [b]' [P] {E} = -[b] [T]+ [(zet)T] [P] (12)The electric variables are connected by the relation (13)[D] = (eps)o * [E] + [P]where (eps)o is the permittivity of free space, defined by (14)(eps)o * (mu)o * (c) * (c) = 1;(mu)o is the permeability of free space, equal, by definition, to $4 * PI * 10^{(-7)}$, and (c) is the velocity of light in vacuo and, also by definition, is equal exactly to 2.99792458 x 10^8 m/s. From (13) the expressions for the remaining electric variables associated, respectively, with the six equation sets (1) to (12) may be found: (15)[P] = [e] [S] + [(chi)S][E] [d] [T] + [(chi)T](16)[P] = [E] (eps)o * [h] [S] + [I - (eps)o * (bet)S] [D] [P] = (17)(18)[P] = (eps)o * [g] [T] + [I - (eps)o * (bet)T] [D]

$$= [I + (zet)T * (eps)o] [g]$$
 (37)

Some alternative relations are the following:

$$[a - h] = [(zet)S] [h] * (eps)o$$

= [(bet)S] [a] * (eps)o (38)

$$[b - g] = [(zet)T] [g] * (eps)o$$

= $[(bet)T] [b] * (eps)o$ (39)

$$[e + a * (eps)o] = [(eps)S] [a]$$
(40)

$$[d + b * (eps)o] = [(eps)T] [b]$$
(41)
(42)

$$[e - h * (eps)o] = [(chi)S] [h]$$
 (42)

$$[d - g * (eps)o] = [(chi)T] [g]$$
(43)

Equations (21) to (43) result from equating like dependent variables pairs selected from equations (1) to (12) and (15) to (20).

Each pair yields one equation in three variables, one mechanical and two electrical, or vice versa. Two other equations exist, again from (1) to (12) and (15) to (20), that contain the same three variables found in each paired equation. One of these auxiliary equations is used to eliminate one of the two variables of the same kind; the result is one equation in two variables, one electrical and one mechanical. These are now independent variables, so the coefficients must vanish; two relations between the material coefficients result. As an example, (3) and (7) both have [S] as dependent variable. Equating them produces one relation in [T], [E], and [D]; one of the electrical variables must be eliminated. This is done by using either (4) or (8); each contains the same three variables. If (8) is used to eliminate [E], one obtains [sE -d'g - sD [T] = [d' (bet)T - g'] [D]. Therefore, [sE] - [sD] = [d]' [g] and [g] = [(bet)T] [d]. Use of (4) instead of (8) leads to the equations [sE] - [sD] = [g]' [d] and [d] = [(eps)T] [g]. There are 36 pairs, six each equating [S] and [T], and eight each equating [E], [D], and [P]. The 72 relations contain many redundancies. Relations between the elastic, piezoelectric, and dielectric constants are shown schematically in Tables 2 and 3.

CALCULATION SEQUENCE

Using as input [sE], [d], and [(eps)T], one may compute the remaining quantities in a variety of ways. The following sequence is typical:

[CE] = [SE] (-1)	(44)
[(bet)T] = [(eps)T] (-1)	(45)
[e] = [d] [cE]	(46)
[(eps)T] - [(eps)S] = [e] [d]'	(47)
[(eps)S] = [(eps)T] - [e] [d]'	(48)
[(bet)S] = [(eps)S] (-1)	(49)
[h] = [(bet)S] [e]	(50)
[cD] - [cE] = [e]' [h]	(51)
[cD] = [cE] + [e]' [h]	(52)
[g] = [(bet)T] [d]	(53)
[sE] - [sD] = [d]' [g]	(54)
[sD] = [sE] - [d]' [g]	(55)
[(betr)S] = [(bet)S] * (eps)o	(56)
[(zetr)S] = [(betr)S] (I - (betr)S] (-1)	(57)



TABLE 2. RELATIONS AMONG MATERIAL CONSTANTS.



TABLE 3. FURTHER RELATIONS AMONG MATERIAL CONSTANTS.

[(zet)S] = [(zetr)] / (eps)o(58) [(betr)T] = [(bet)T] * (eps)o(59)[(zetr)T] = [(betrT] [I - (betr)T] (-1)](60) [(zet)T = [(zetr)T] / (eps)o(61) $[(chi)S] = [(zet)S]^{(-1)}$ (62) $[(chi)T] = [(zet)T]^{(-1)}$ (63)[a] = [(zet)S] [e](64) [b] = [(zet)T] [d](65) [CP] - [CE] = [e]' [a](66) [CP] = [CE] + [e]' [a](67) [CP] - [CD] = [a]' [h] * (eps)o(68) [sE] - [sP] = [d]' [b](69) [sP] = [sE] - [d]' [b](70)[sD] - [sP] = [g]' [b] * (eps)o(71)[(bet)S] - [(bet)T] = [h] [g]'(72) [(chi)T] - [(chi)S] = [(eps)T] - [(eps)S](73) [(zet)S] - [(zet)T] = [a] [b]'

A number of these relations are used as checks. For example, [(bet)S] and [(bet)T] are known from (45) and (49), but the difference is recomputed in (72).

(74)

EXPLICIT FORMULAS FOR POINT GROUP 4mm

Elastic:

The 6x6 elastic constant portion of Table 4 partitions into 4x4 and 2x2 submatrices. The 4x4 elastic stiffness and compliance submatrices are interrelated by formulas (75) to (93). The elastopiezodielectric matrix for class 4mm is found in Cady (Ref. 2). Other references to lithium tetraborate are given in Refs. 3 to 26.

A = s33 * (s11 + s12) - 2 * s13 * s13(75)B = (s11 - s12)(76)cll = +(sl1 * s33 - sl3 * sl3) / (A * B)(77)

TABLE	4.	ELAST	OPIEZO	DIEL	ECTRIC	: M	ATRICES	5 FOR	POINT	GROUP	4mm.
11	12	13	00	00	00]	00	00	31		
12	11	13	00	00	00]	00	00	31	CE]]-	
13	13	33	00	00	00	j	00	00	33	C]((cp3)0
00	00	00	44	00	00	j	00	15	00		h (
00	00	00	00	44	00]]	15	00	00]-	
00	00	00	00	00	66]	00	00	00	n jo	Del)S
00	00	00	00	15	00]	11	00	00	aD 1	. 1
00	00	00	15	00	00]]	00	11	00]-	
31	31	33	00	00	00]	00	00	33	a](2013

Matrix entries show only subscripts.

c12 = -(s12 * s33 - s13 * s13) / (A * B)(78)c13 = - s13 / A(79) $c_{33} = (s_{11} + s_{12}) / A$ (80) c44 = 1 / s44(81) c66 = (c11 - c12) / 2 = s44 / (2 * B)(82) K = c33 * (c11 + c12) - 2 * c13 * c13(83) L = (c11 - c12)(84)sll = +(cll * c33 - cl3 * cl3) / (K * L)(85) s12 = -(c12 * c33 - c13 * c13) / (K * L)(86) s13 = - c13 / K(87) $s_{33} = (c_{11} + c_{12}) / K$ (88) s44 = 1 / c44(89)s66 = 1 / c66(90) det (3x3) [s] = A * B (91) det (3x3) [c] = K * L (92) A * K = B * L = A * B * K * L = 1(93) Formulas (75) to (93) hold for each set of constant electrical conditions: either E, D, or P constant. [CD] - [CE] = [del CDE] = [e]' [h] = [h]' [e](23)del cDE11 = + e31 h31(94) del cDE12 = + e31 h31(95) del cDE13 = + e31 h33 = + h31 e33 (96) del cDE33 = + e33 h33(97) del cDE44 = + e15 h15(98) del cDE66 = 0(99) [CP] - [CD] = [del CPD] = [a]' [h] * (eps)o

= [h]' [a] * (eps)o (24)

del cPD11 = (+ a31 h31) * (eps)o(100)del cPD12 = (+ a31 h31) * (eps)o(101) del cPD13 = (+ a31 h33) * (eps)o(102) = (+ h31 a33) * (eps)odel cPD33 = (+ a33 h33) * (eps)o(103)del cPD44 = (+ a15 h15) * (eps)o(104)(105)del cPD66 = 0(25) [CP] - [CE] = [del CPE] = [e]' [a] = [a]' [e]del cPE11 = + e31 a31(106) del cPE12 = + e31 a31(107) del cPE13 = + e31 a33 = + a31 e33 (108) del cPE33 = + e33 a33 (109) del cPE44 = + e15 a15 (110)del cPE66 = 0(111)From the del c13 entries we have the ratios $e_{31} / e_{33} = h_{31} / h_{33} = a_{31} / a_{33}$. (112)[sE] - [sD] = [del sED] = [d]' [g] = [g]' [d](26) del sED11 = + d31 g31 (113) del sED12 = + d31 g31 (114) del sED13 = + d31 g33 = + g31 d33 (115)(116) del sED33 = + d33 g33 del sED44 = + d15 g15(117) del sED66 = 0(118)[sD] - [sP] = [g]' [b] * (eps)o= [b]' [g] * (eps)o(27) del sDP11 = (+ g31 b31) * (eps)o (119)

del sDP12 = $(+ \alpha 31 \ b 31) \ * (ens) \alpha$	(120)
$dol \ sDP13 = (+ a31 \ b33) \ \star (eps) 0$	(,
$\frac{1}{2} = (+ \frac{1}{2}) = (+ \frac{1}{2}) = (-\frac{1}{2}) = (-$	(121)
= (+ b31 g33) = (eps)0	(122)
del $SDP33 = (+ g33 b33) * (eps)0$	(122)
del sDP44 = (+ g15 b15) * (eps)0	(123)
del sDP66 = 0	(124)
[sE] - [sP] = [del sEP] = [b]' [d] = [d]' [b]	(28)
del sEP11 = + d31 b31	(125)
del sEP12 = + d31 b31	(126)
del sEP13 = + d31 b33 = + b31 d33	(127)
del sEP33 = + d33 b33	(128)
del sEP44 = + d15 b15	(129)
del sEP66 = 0	(130)
From the del s13 entries we have the ratios	
d31 / d33 = g31 / g33 = b31 / b33.	(131)
Piezoelectric:	
[e] = [d] [cE]	(33)
e15 = + d15 cE44	(132)
e31 = + d31 (cE11 + cE12) + d33 cE13	(133)
e33 = + d33 cE33 + d13 cE13 * 2	(134)
[h] = [(bet)S] [e]	(34)
h15 = (bet)S11 e15	(135)
h31 = (bet)S33 = 31	(136)
h33 = (bet)S33 e33	(137)
[g] = [(bet)T] [d]	(35)
q15 = (bet)T11 d15	(138)
a31 = (bet)T33 d31	(139)
q33 = (bet)T33 d33	(140)

.

[a] = [(zet)S] [e]	(36)
a15 = (zet)S11 e15	(141)
a31 = (zet)S33 e31	(142)
a33 = (zet)S33 e31	(143)
[b] = [(zet)T] [d]	(37)
b15 = (zet)T11 d15	(144)
b31 = (zet)T33 d31	(145)
b33 = (zet)T33 d33	(146)

Dielectric:

$$[(bet)Y] = [(eps)Y]^{(-1)}$$
(21)

$$(het) Y_{11} = 1 / (eps) Y_{11}$$
 (147)

$$(bet) Y_{33} = 1 / (eps) Y_{33}$$
 (148)

$$[(zetr)Y] = [(betr)Y] [I - (betr)Y]^{(-1)}$$
(149)

$$(zet)Y11 = 1 / ((eps)Y11 - (eps)o)$$
 (150)

$$(-2)^{12} = 1 (((2)^{2})^{2} = (2)$$

$$(151)$$

$$(zet)YII = I / ((eps)YII - (eps))$$
(151)

$$(eps) Y_{33} = 1 / ((eps) Y_{33} - (eps) o)$$
(151)

(30)

(152)

(153)

(31)

(154)

(155)

(156)

(157)

(158)

$$(2ec)^{111} - 1 / ((eps)^{111} (eps)^{0})$$
 (151)

$$(zet)$$
 V33 = 1 / ((eps) V33 - (eps) o) (151)

$$(2et)^{11} = 1 / ((eps)^{11} (eps)^{0})$$
 (151

[(eps)T - (eps)S] = [del (eps)] = [e] [d]' =

del (eps)11 = del (chi)11 = + e15 d15

[(bet)S - (bet)T] = [h] [g]' = [g] [h]'

[(chi)T - (chi)S] = [del (chi)] = [d] [e]'

del (eps)33 = del (chi)33 = + e33 d33 + e31 d31 * 2

del (bet)11 = + h15 g15

[(zet)S - (zet)T] = [del (zet)] = [a] [b]' = [b] [a]'

del (zet)11 = + a15 b15

del (bet)33 = + h33 g33 + h31 g31 * 2

del (zet)33 = + a33 b33 + a31 b31 * 2

$$zet Y_{33} = 1 / ((eps)Y_{33} - (eps)o)$$
(151)

$$(2et)^{11} = 1 / ((eps)^{11} (eps)_{0})$$
 (151)

$$(2et) V_{33} = 1 / ((eps) V_{33} - (eps) o)$$
 (151)

$$(zet)$$
 YII = 1 / ((eps) YII - (eps) 0) (150)

$$zet Y_{33} = 1 / ((eps)Y_{33} - (eps)o)$$
(151)

$$zet Y11 = 1 / ((eps)Y11 - (eps)0)$$
(15)

$$zet$$
) Y11 = 1 / ((eps) Y11 - (eps) 0) (150)

$$zet)Y11 = 1 / ((eps)Y11 - (eps)0)$$
 (150)

$$zet)Y11 = 1 / ((eps)Y11 - (eps)o)$$
 (150

$$(zet)^{11} = 1 / ((eps)^{11} (eps)^{0})$$
 (151)

$$(zet)YII = I / ((eps)YII - (eps)o)$$
 (150)

$$(25)$$
 $((eps))$ $((eps))$ $((eps))$ (15)

$$zet)$$
 Y11 = 1 / ((eps) Y11 - (eps) 0) (151

$$(2et)$$
 YII = 1 / ((eps) YII - (eps) 0) (150)

$$(zet)Y_{33} = 1 / ((eps)Y_{33} - (eps)o)$$
 (151)

INPUT VALUES FOR LI2 B4 07

The values measured by Shiosaki, et al. (Ref. 1) are as follows:

 TABLE 5. ISAGRIC ELASTIC COMPLIANCES.

 sE11
 sE12
 sE13
 cE33
 sE44
 sE66

 8.81
 1.23
 -5.92
 24.6
 17.1
 21.4

 Units:
 10⁽⁻¹²⁾ m/N.

 TABLE
 6. PIEZOELECTRIC STRAIN COEFFICIENTS.

 d15
 d31
 d33

 d15
 c31
 d33

 d15
 c31
 d33

 d15
 c31
 d33

 d15
 c31
 d34

 units:
 10⁽⁻¹²⁾ m/y.
 m/y.

TABLE 7. DIELECTRIC PERMITTIVITIES AT CONSTANT STRESS. (eps)T11 (eps)T33 (eps)T11/(eps)o (eps)T33/(eps)o 82.61 87.92 9.33 9.93 Units: 10⁽⁻¹²⁾ F/m. F/m.

OUTPUT VALUES FOR LI2 B4 07

The input values from Tables 5, 6, and 7 were used to compute the remaining elastic, piezoelectric, and dielectric quantities for lithium tetraborate in the manner discussed in prior sections of this report. The results are given in Tables 8 to 15.

	=======================================					
	cE	cD	cP	del CDE	del cPE	del cPD
					============	1232222222
11	135.5	136.7	136.8	1.18	1.35	0.167
12	3.57	4.75	4.92	1.18	1.35	0.167
13	33.47	37.24	37.78	3.78	4.31	0.535
33	56.76	68.83	70.54	12.07	13.78	1.71
44	58.48	61.31	61.66	2.83	3.18	0.358
66	46.73	46.73	46.73	0.0	0.0	0.0
Units:	10(9) _{N/r}	n2.				

TABLE	8.	ELASTIC	STIFFNESSES.

TABLE 9. ELASTIC COMPLIANCES.

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	===========			=================		===========
	sE	sD	sP	del sED	del sEP	del sDP
=====					===========	*==*****
11	0 01	0 7 7	0 70	0 0757	0.0040	0 00040
10	0.01	8./3	8.73	0.0757	0.0842	0.00848
12	1.23	1.15	1.15	0.0757	0.0842	0.00848
13	-5.92	-5.30	-5.29	-0.569	-0.633	-0.0637
33	24.6	20.3	19.8	4.28	4.76	0.479
44	17.1	16.3	16.2	0.788	0.883	0.0946
66	24.4	21.4	21.4	0.0	0.0	0.0
Units:	10(-12) m	2/N.				
TABLE 10. PIEZOELECTRIC [e], [h], AND [a] VALUES.						
	e======== e		============== h		a=====================================	
	===========					
15	0.4	472	5.9	9	6.75	
31	0.3	290	4.0	7	4.64	
33	0.9	928	13.0	0	14.84	
					_	
Units:	e: C/m2	; h and a	: 10(9)	V/m.		
		==========	~~~~~~~			

TABLE 11.	PIEZOELECTRI	C [d], [g], AND	[b] VALUES.		
	d	g	b		
15 31 33	8.07 -2.58 19.4	97.7 -29.3 220.6	109.4 -32.6 245.4		
Units: d:	10 ⁽⁻¹²⁾ m/V	; g and b: 10(-	³⁾ m2/C.		
TABLE 12.	DIELECTRIC (eps) VALUES.			
============	(eps)S	(eps)T	del (eps)TS	=====	
11 33	78.80 71.41	82.61 87.92	3.81 16.51		
Units: 10	(-12) F/m.				
del (eps)T	5 = del (chi)'	 ГS			
TABLE 13. DIELECTRIC (chi) VALUES.					
	(chi)S	(chi)T	del (chi)TS	=====	
11 33	69.95 62.56	73.76 79.07	3.81 16.51		
Units: 10	(-12) _{F/m} . ==============			=====	
del (chi)TS	S = del (eps)	ſS			
TABLE 14.	DIELECTRIC ()	pet) VALUES.			
	(bet)S	(bet)T	del (bet)TS	=====	
11 33	12.69 14.00	12.11 11.37	-0.585 -2.63		
Units: 10	(9) _{m/F} .				

TABLE 15	BLE 15. DIELECTRIC (zet) VALU			
	(zet)S	(zet)T	del (zet)TS	
11 33	14.30 15.99	13.56 12.65	-0.738 -3.34	
Units: ========	10 ⁽⁹⁾ m/F.			

CONCLUSIONS

This report provides formulas interrelating the coefficients that appear in the several alternative sets of constitutive equations involving the elastic, piezoelectric, and dielectric properties of crystals. These are then specialized for crystals of class 4mm; using measured values reported for lithium tetraborate, numerical values of the elements of the polarization matrices are calculated.

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