Correction of elastic, piezoelectric and dielectric constants of $NdCa_4O(BO_3)_3$ crystal using measured SAW parameters

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INTRODUCTION

Recently there is an increasing need for devices and sensors that could work in harsh environments at elevated temperatures. A lot of investigations were made over this problem. One of the possible solution is using piezoelectric transducers with surface acoustic waves (SAW). Performance of such a transducers is related with electrodes deposited on piezoelectric substrate crystal and with crystal's physical properties. Gallium ortophosphate ($GaPO_4$) can be used up to 930°C but above this temperature there is no piezoelectric effect due to phase transition [1]. Langasite ($La_3Ga_5SiO_{14}$ abbrev: LGS), new piezoelectric material shows no phase transitions up to the melting point (1500°C) but its electrical resistivity falls down with temperature and limits application ability to 800°C [2]. A neodymium calcium oxyborate crystal belonging to *Re*COB family (where *Re*= Gd, Nd, Y, La) [3] shows no phase transition and holds piezoelectricity up to the melting point ~1470°C. For YX orientation it is thermally compensated around room temperature as good as ST-cut quartz and shows electromechanical coupling coefficient 0.7%. This coupling coefficient is six times higher than for ST-cut quartz and the highest all over known *Re*COB. On the other hand, for ZX and XZ orientations, NdCOB is thermally sensitive and its temperature coefficient of frequency is about -80 ppm/°C [4]. These properties make NdCOB crystal attractive for SAW components and high temperature sensor applications. In SAW component design procedure it is important to precisely simulate crystal's SAW properties. For SAW simulations, a set of material constants is needed. NdCOB belongs to monoclinic class, point group *m* that is described by 13 elastic s^E_{IJ}, 10 piezoelectric d_{iJ} and 4 dielectric ε^S_{ij} constants (Fig.1), where I, J = 1, 2..6; i, j = 1, 2, 3; superscripts E and S indicate constant or zero electric field and deformation, respectively (will be skipped in fallow consideration). These 27 constants were previously measured by T. Karaki, M. Adachi, Y. Kuniyoshi [5] by resonance-antiresonance method. Using these constants in SAW simulations lead to results that are significantly different than measurements. The difference is caused by material constants errors generated by measurements of large number of specimens differently oriented relative to crystal axes. So the material constants correction would be desirable. A material constants correction based on SAW velocity measurements for simple, strong piezoelectric crystal like lithium niobate (6 elastic constants, 4 piezoelectric and 2 dielectric) was proposed in [6]. We propose a material constants correction based on measured SAW velocity and electromechanical coupling coefficient. Additionally dielectric constants evaluation based on effective permittivity measurements will be made. All the measurements will be made by using SAW delay line.

$$s_{IJ} = \begin{bmatrix} s_{11} & s_{12} & s_{13} & 0 & s_{15} & 0 \\ & s_{22} & s_{23} & 0 & s_{25} & 0 \\ & & s_{33} & 0 & s_{35} & 0 \\ & & & s_{44} & 0 & s_{46} \\ & & & & s_{55} & 0 \\ & & & & & s_{66} \end{bmatrix} \text{a)}$$

$$d_{iJ} = \begin{bmatrix} d_{11} & d_{12} & d_{13} & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & d_{26} \\ d_{31} & d_{32} & d_{33} & 0 & d_{35} & 0 \end{bmatrix} \text{b)}$$

$$\varepsilon_{ij} = \begin{bmatrix} \varepsilon_{11} & 0 & \varepsilon_{13} \\ & \varepsilon_{22} & 0 \\ & & \varepsilon_{33} \end{bmatrix} \text{c)}$$

Fig. 1 Material constants matrices: elastic s^E_{IJ} -a), piezoelectric d_{iJ} -b) and dielectric ε^S_{ij} -c)

CALCULATIONS AND EXPERIMENTS RESULTS

In monoclinic *m* class a rectangular (X, Y, Z) coordinate system relative to crystallographic (a, b, c) system is chosen such that Y||b , Z||c , X axis is perpendicular to Y and Z axes and forms right-handed system. For finding SAW propagation directions where velocity is most sensitive on to elastic constants changes, a computer simulations were made in three planes perpendicular to X, Y and Z axes. Velocity changes Δv were calculated for elastic constants changes of about $\pm 1\%$ versus propagation angle α (Fig.2). For X-cut α is taken between SAW propagation direction and Y axis, for Y and Z cuts, α is taken between SAW direction and X axis. For SAW velocity calculation, NdCOB constants from [5] and J. Campbell, R. Jones algorithm [7] were used.

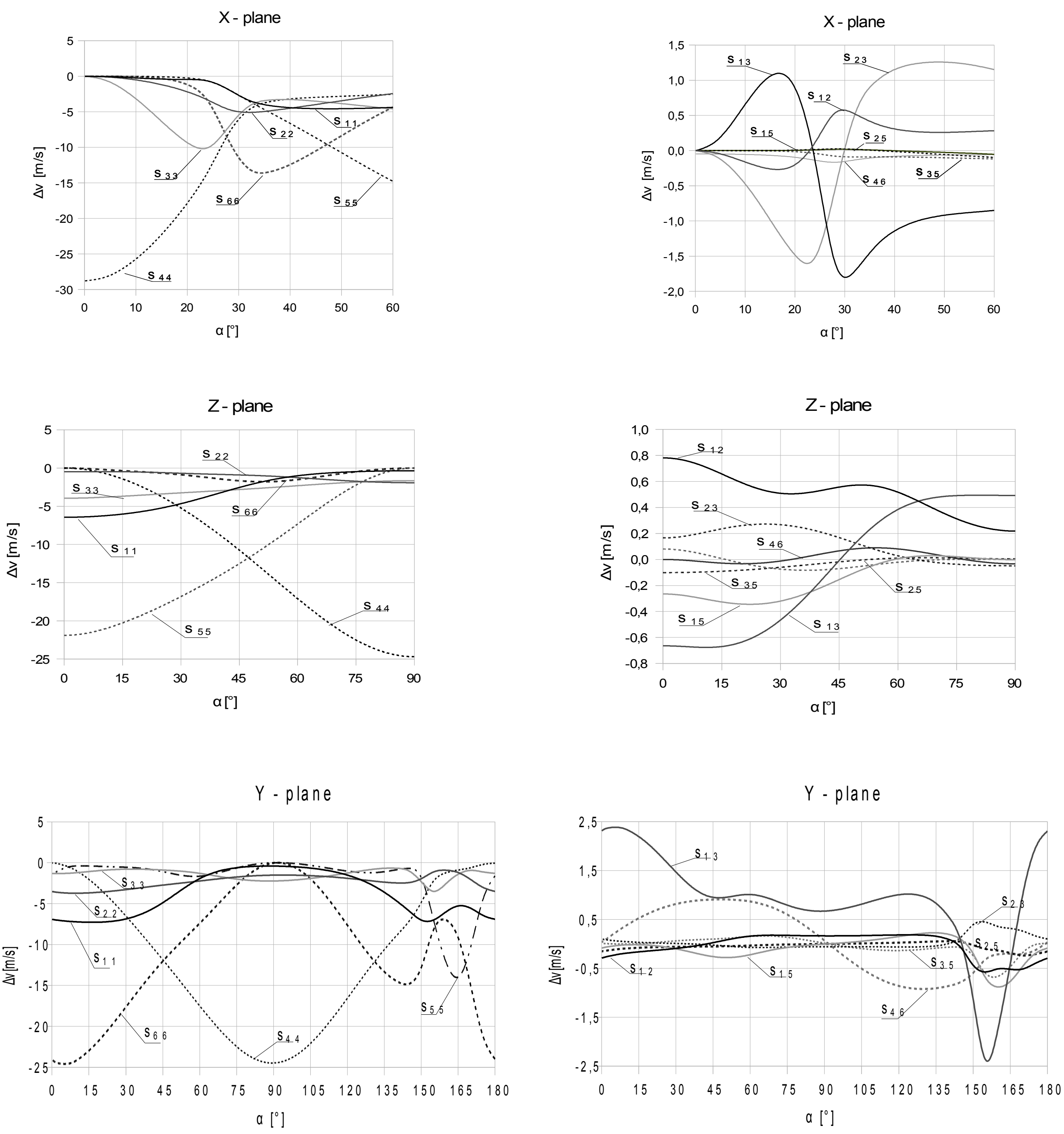

Fig. 2. Velocity changes induced by $\pm 1\%$ s_{IJ} change versus propagation angle for X, Y and Z planes.

For all planes the velocity is most sensitive on to s_{44} and s_{55}. For X-plane, s_{11}, s_{22}, s_{33}, s_{66} show good sensitivity while s_{13}, s_{12} and s_{23}, show low but acceptable sensitivity. For Z-plane, $s_{11}$and s_{33} show medium sensitivity. For Y-plane, s_{11}, s_{22}, s_{66} show good sensitivity while s_{13} shows low sensitivity. Elastic constants s_{15}, s_{25}, s_{35}, s_{46} induced very low Δv. This indicate that these 4 constants can not be corrected by using SAW. So 9 of 13 elastic constants will be corrected on the basis of 9 orientation measurements. For dielectric constants it was found that d_{11} gives low electomechanical coupling change. So 9 of 10 dielectric constants will be corrected.

Measurement method and results

The crystal was grown by Cz. technique along crystallographic b axis in iridium crucible at nitrogen atmosphere. For compound powder preparation a 5N pure Nd_2O_3, 4N pure $CaCO_3$ and B_2O_3 with less than 200 ppm water condensation were used. A SAW delay line with double-electrode interdigital transducers (IDT) was designed [4] for measure effective permittivity ε_e, SAW properties. An aluminium layer of about 0.1 µm was used for delay line fabrication by the lift-off method. Amplitude responses were measured in the 50Ω system (Agilent Technologies network analyser type 8753ET). As an example, the measured amplitude response for YX10° orientation is shown in Fig.3.

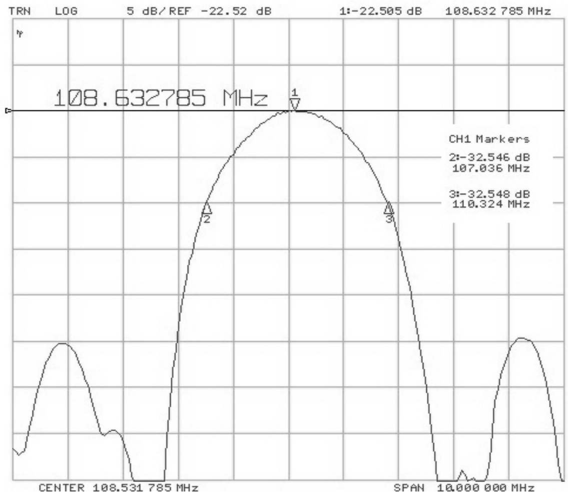

Fig.3 Measured amplitude response of delay line for YX10° oriented NdCOB substrate

Delay line's capacitances needed for ε_e evaluation were measured by bridge (Marconi Instruments) for XZ, YZ, YX and YX40^0 orientation, ε_e was then evaluated from [9]:

$$C_p = N \cdot 0.707 \cdot W(\varepsilon_0 + \varepsilon_e) \tag{1}$$

where C_p is transducer's capacitance, N is the number of electrodes, ε_0 is the dielectric constant of the vacuum, W is transducer's aperture. Obtained ε_e are shown in Tab.1.

Table 1. Measured effective permittivities

orientation	$\varepsilon_e/\varepsilon_0$
YX	11,9
YX40°	10,7
YZ	12
XZ	9,5

Dielectric constants (Tab.2) were obtained from:

$$\varepsilon_e = \sqrt{\varepsilon_{11}' \cdot \varepsilon_{33}' + \varepsilon_{13}'} \tag{2}$$

where ε'_{ij} are dielectric constants in coordinate system related with SAW propagation direction.

Table 2. Initial and measured dielectric constants

$\varepsilon_{ij}/\varepsilon_o$	initial [1]	measured	$\Delta\varepsilon_{ij}/\varepsilon_{ij}^0$ [%]
1 1	9,9	9,9	0
1 3	-0,8	-1,9	137,5
2 2	15	15,2	1,33
3 3	10	9,5	-5

SAW free velocity v_f and electromechanical coupling coefficient K^2 were evaluated by comparing measured and calculated delay lines amplitude characteristics [8]. Evaluated v_f and K^2 are shown in Tab.3.

Table 3. Measured SAW parameters

n	orientation	$\varepsilon_e/\varepsilon_0$	v_f	K^2
---	---	---	m/s	%
1	XY20°	10,3	3160	0,13
2	XY30°	10,5	3334	1,1
3	YX	11,9	3510	0,7
4	YX10°	11,5	3483	0,5
5	YX20°	11,1	3377	0,44
6	YX40°	10,7	3180	0,3
7	YZ	12	2786	0,08
8	ZX	10,6	3239	0,27
9	ZY	11,8	2780	0,08

Correction procedure

Correction procedure's first step (Fig.4) is to assume density and initial values of elastic, piezoelectric and dielectric constants. Then orientations with SAW velocity most sensitive on to elastic constants change should be evaluated. The number of chosen orientations should be at least equal to number of elastic constants that will be corrected. Then delay lines should be deposited on such oriented crystal substrates. Next step is to determine effective permittivities by measuring delay line's capacitances. Then dielectric constants could be determined from effective permittivities. Next step is to determine SAW velocities and electromechanical coupling coefficients from delay line's amplitude characteristics. Then calculated SAW velocities should be fitted with measured ones using least squares algorithm. As the target function a sum of squares of velocities (*VSQ*) should be taken:

$$VSQ = \sum_n (v_c[n] - v_m[n])^2 \quad (3)$$

where v_c and v_m are calculated and measured SAW free velocities, respectively, n is the orientation number. *VSQ* should be minimized by elastic constants modification while piezoelectric constants stay unchanged at this stage. In the next step calculated and measured electromechanical coupling coefficients should be fitted by piezoelectric constants modification. This time elastic constants, obtained at previous step remain unchanged. As the target function a sum of squares of electromechanical coupling coefficients (*KSQ*) should be taken:

$$KSQ = \sum_n (K_c^2[n] - K_m^2[n])^2 \quad (4)$$

where K^2_c and K^2_m are calculated and measured SAW electromechanical coupling coefficients, respectively, where:

$$K^2 = 2\cdot(v_f - v_s)/v_f \quad (5)$$

where v_f and v_s are SAW velocities under free and electrically shorted surface condition, respectively. The minimization procedures are repeated until both *VSQ* and *KSQ* reach its minimal values.

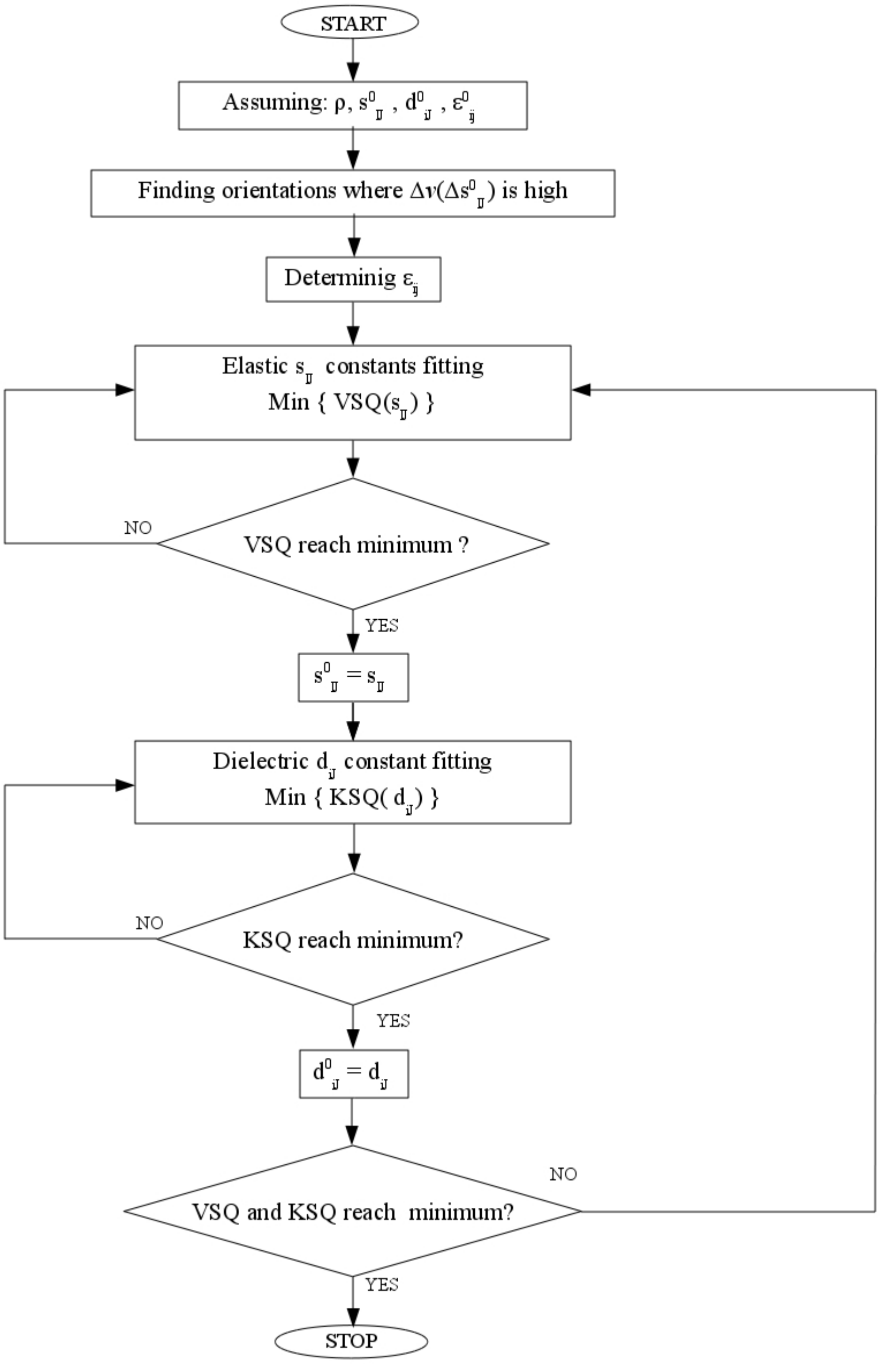

Fig.4. Correction procedure

For *VSQ* and *VSK* function minimization a computer program was written that uses a R. Hooke, T. Jeeve's algorithm [10]. It checks local function behavior and then make a step in to function's minimum direction. It works without gradients so it is applicable to wide range of functions.

Correction results

Fitted elastic and dielectric constants are shown in Tab.4 and Tab.5 respectively.

Table 4. Initial and corrected elastic constants

s_{IJ}	Initial [5]	corrected	$\Delta s_{IJ}/s_{IJ}^{o}$
---	$[10^{-12} m/N]$	$[10^{-12} m/N]$	[%]
1 1	8,3	9,3135	12,21
1 2	-2	-0,0420	-97,90
1 3	-3,5	-3,1000	-11,43
1 5	-0,9	-0,9	0
2 2	7,5	7,7855	3,81
2 3	-1,6	-3,0661	91,63
2 5	0,5	0,5	0
3 3	9,4	8,4132	-10,50
3 5	0,9	0,9	0
4 4	34	33,873	-0,37
4 6	1	1	0
5 5	22	22,368	1,67
6 6	20	18,889	-5,56

Table 5. Initial and corrected dielectric constants

d_{iJ}	Initial [5]	corrected	$\Delta d_{iJ}/d_{iJ}^{o}$
---	$[10^{-12}$ C/N]	$[10^{-12}$ C/N]	[%]
1 1	1,7	1,7	0
1 2	3,9	2,6788	-45,59
1 3	-4,9	-4,8735	-0,54
1 5	3	3,5281	14,97
2 4	4,5	2,9452	-52,79
2 6	16,5	17,7770	7,18
3 1	-1,4	-0,4321	-224
3 2	-2,5	3,5449	170,52
3 3	1,5	2,3857	37,13
3 5	5,7	4,1321	-37,94

Differences between measured and calculated SAW parameters using initial and corrected constants are shown in Tab.6 and Tab.7.

Table 6. Differences between measured and calculated SAW velocity

orientation	Δv [m/s]	Δv [m/s]
---	for initial constants	for corrected constants
XY20	10	0,6
XY30	-38	2,6
YX	53	2
YX10	-71	3,9
YX20	2	4,3
YX40	-17	-3,8
YZ	-2	-0,2
ZX	26	1,7
ZY	4	4,8

Table 7. Differences between measured and calculated K^2

orientation	ΔK^2 [%]	ΔK^2 [%]
---	for initial constants	for corrected constants
XY20	9,4	-2,5
XY30	-5,7	-6,3
YX	6,4	-10,3
YX10	18	4,6
YX20	7,36	-2,5
YX40	-8	-7,5
YZ	-7,6	-9,2
ZX	-23	-6,8

Conclusions

For dielectric constants, only ε_{13} was significantly changed during correction prosess. This is because ε_{13} is small and have low influence on to capacitance. Mostly changed elastic constants are s_{12} and s_{23}. Such a big change is due to low accuracy for these constants in resonance-antiresonance method. A mean for other constants change is about 6 %. Mostly changed piezoelectric constants are d_{31} and d_{32}. Differences between measured and calculated SAW parameters are lower for corrected material constants.

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