

Elastic, Piezoelectric, and Dielectric Properties of $0.955\text{Pb}(\text{Zn}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-}0.45\text{PbTiO}_3$ Single Crystal with Designed Multidomains

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Abstract—The elastic, piezoelectric, and dielectric properties of a $0.955\text{Pb}(\text{Zn}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-}0.045\text{PbTiO}_3$ (PZN-4.5%PT) multi-domain single crystal, poled along [001] of the original cubic direction, have been determined experimentally using combined resonance and ultrasonic methods. At room temperature, the PZN-4.5%PT single crystal has rhombohedral symmetry. After being poled along [001], four degenerate states still remain. Statistically, such a domain-engineered crystal may be treated as having an average tetragonal symmetry, and its material constants were determined based on 4 mm symmetry. It was confirmed that the electromechanical coupling coefficient k_{33} for the domain-engineered samples is $>90\%$, and the piezoelectric constant d_{33} is >2000 pC/N. A soft shear mode with a velocity of 700 m/s was found in the [110] direction. From the measured experimental data, the orientational dependence of phase velocities and electromechanical coupling coefficients was calculated. The results showed that the transverse and longitudinal coupling coefficients, k_{31} and k_{33} , reach their maximum along [110] and [001], respectively.

I. INTRODUCTION

ALTHOUGH early work on $\text{Pb}(\text{Zn}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$ (PZN-PT) solid solution system can be dated back to the 1960s [1], [2], it was not until very recently that this relaxor-based ferroelectric single crystal system has generated a great deal of attention. It was found that the single crystal system exhibits an extraordinary large electromechanical coupling coefficient k_{33} ($>90\%$) and piezoelectric coefficient d_{33} (>2000 pC/N) at room temperature after being poled in [001], which is off the polarization direction of $\langle 111 \rangle$ [3]–[5]. Considering that the best piezoelectric materials used today, the modified $\text{Pb}(\text{Zr}_x\text{Ti}_{1-x})\text{O}_3$ (PZT), only has k_{33} of 75% and d_{33} of 700 pC/N, the new system shows a promising potential to produce higher sensitivity ultrasonic transducers with superior broadband characteristics [6], large strain actuators, and other electromechanical devices.

The physical mechanism of this multidomain system is still unclear at the moment. The lack of complete physical property data is the main hindrance for theoretical studies.

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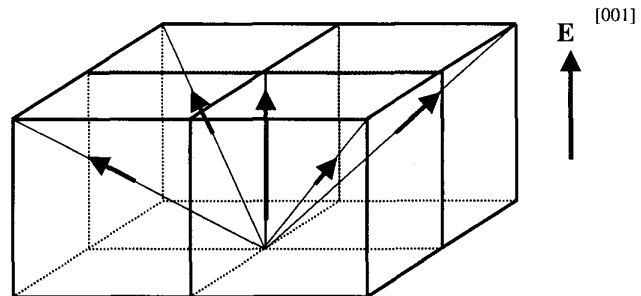


Fig. 1. Possible domain configuration and associated polarization orientation in a PZN-4.5%PT single crystal after being poled in [001] direction.

Practically, it is also very important to have a complete set of elastic, piezoelectric, and dielectric constants available. At present, only one electromechanical coupling coefficient k_{33} and one piezoelectric coefficient d_{33} have been reported [3]–[5]. The aim of this paper is to provide a complete set of such data for the PZN-4.5%PT domain-engineered single crystal. These data were obtained by using a combined method involving both pulse-echo and impedance resonance techniques.

Above 150°C , the crystal PZN-4.5%PT has a cubic perovskite structure with symmetry $m\bar{3}m$ but becomes a rhombohedral ferroelectric phase with 3m symmetry below the phase transition temperature [5]. When an electric poling field along [001] of the original cubic axis is applied to the crystal, a multidomain configuration can be produced (as shown in Fig. 1) consisting of four degenerate states and charged domain walls [5]. For convenience, the coordinate systems used for the orientations in this paper are all based on the high temperature cubic phase. Statistically, the poled system should have a pseudo-tetragonal 4 mm macroscopic symmetry, although the microscopic symmetry of the system is 3m [5]. Our measured material properties presented in this paper are based on this 4 mm symmetry. There are total of 11 independent physical constants describing the elasto-piezo-dielectric matrices for the 4 mm symmetry [7].

The dielectric constants ϵ_{11}^T and ϵ_{33}^T were calculated from the low frequency capacitance using the parallel capacitor approximation. Two of the elastic compliances, s_{11}^E and s_{33}^D , and the two electromechanical coupling coefficients, k_{33} and k_{31} , were calculated from the resonance and antiresonance frequencies of the length-extensional modes

of long vibration bars. Another five elastic constants (c_{11}^E , c_{12}^E , c_{44}^E , c_{66}^E , and c_{44}^D) were determined from the measurements of phase velocities of ultrasonic waves propagating along certain orientations. Using this measurement scheme, the only samples needed are those with the orientations of [001]/[010]/[100] and [001]/[110]/[1 $\bar{1}$ 0]. Thus, fewer samples are required when using this combined measurement technique [8].

For each given wave propagation direction, the relationship between the phase velocity and associated material constants can be obtained by solving the Christoffel wave equations [9], and these velocities can be measured using the pulse-echo technique [7].

II. EXPERIMENTAL PROCEDURE

The impedance resonance method and ultrasonic pulse-echo technique, described in the IEEE standards on piezoelectricity [7], are often used to characterize material properties for piezoelectric materials [10], [11]. Theoretically, all independent elastic, piezoelectric, and dielectric constants for any crystal symmetry can be determined either by the resonance method or by the ultrasonic method, provided there are sufficient numbers of differently oriented samples. In reality, however, for materials of lower symmetry, some geometries for resonance measurements are difficult to prepare, especially when the available crystal is too small to make large aspect ratio resonators. On the other hand, because the ultrasonic technique can only measure certain elastic constants through the measurement of phase velocities, large errors can be introduced for derived material constants that are not related to pure modes. High acoustic attenuation of certain modes may also damp the propagation of ultrasonic waves in certain directions so that a complete set of material constants for low symmetry systems is also difficult to obtain using the ultrasonic method alone. In this work, we have selected only those simple pure modes in each method and combine the measured data to minimize errors in the results.

In the ferroelectric phase, the dipole in each unit cell of the PZN-4.5%PT crystal is along one of the eight $\langle 111 \rangle$ directions. It has been shown experimentally that the crystal can hold a macroscopic polarization only when the electric field is applied along one of the six $\langle 100 \rangle$ directions [3], [4]. After the application of a poling electric field along [001], there are still four remaining degenerate dipole orientations. Such a poling field creates a multidomain structure with strong elastic interaction among the existing domains. Statistically, the four remaining domains have an equal possibility to form so that the global macroscopic symmetry has been assumed as 4 mm in all previous studies [5].

The as-grown crystals were first orientated using the Laue method with an accuracy of $\pm 0.5^\circ$. Then, each sample was cut and polished into a rectangular parallelepiped shape with three pairs of parallel surfaces. Gold electrodes were sputtered on to the [001] and [00 $\bar{1}$] faces of each sam-

ple, and an external electric field of 1.0 to 1.5 MV/m was applied at room temperature to pole the sample. The final dimensions of the samples used for the ultrasonic measurements were about $3 \times 3 \times 1$ mm³. For the length-extensional resonance measurements, the aspect ratio of the sample exceeded 5:1 to yield nearly pure resonance modes [7]. Several sets of samples were prepared for a consistency check. For the bar samples used in the resonance measurements, different aspect ratios were fabricated to ensure purity of the modes. In addition to samples with [001]/[010]/[100] and [001]/[110]/[1 $\bar{1}$ 0] orientations, a resonance bar with its long dimension in [110] and thickness in [001] was also made to verify the anisotropy calculations.

A 15-MHz longitudinal wave transducer (Ultran Laboratories, Inc., Boalsburg, PA) and a 20-MHz shear wave transducer (Panametrics, Waltham, MA) were used for the pulse-echo measurements. The electric pulses used to excite the transducer were generated by a Panametrics 200-MHz pulser/receiver, and the time of flight between echoes was measured by using a Tektronix 460A digital oscilloscope.

For the length-extensional resonance measurements, a HP 4194A impedance/gain-phase analyzer was employed. The resonance and antiresonance frequencies corresponding to the minimum and maximum values of the impedance-frequency spectrum were obtained and used to calculate the corresponding electromechanical coupling coefficients and elastic compliance.

The dielectric measurements were carried out at 1 kHz using a Stanford Research System SR715 LCR Meter.

III. RESULTS AND ANALYSES

Table I lists the measured phase velocities of the longitudinal and shear waves propagating along different crystal orientations in the domain engineered PZN-4.5%PT single crystals. The measured longitudinal electromechanical coupling coefficient k_{33} , the transverse coupling coefficient k_{31} in [110] and [100] directions, and the frequency constants $2lf_r$ and $2lf_a$ for the resonance bars are given in Table II. Here l is the length of the resonance bar, and f_r and f_a are the resonance and antiresonance frequencies, respectively, obtained from the electrical impedance spectrum. The lateral coupling coefficient k_{31} is different when the long dimension is along [001] or [110] for this system because of the anisotropy of the crystal. For comparison, the coupling coefficients and the frequency constants of PZT-5H, BaTiO₃ ceramic, and BaTiO₃ crystal are also listed in Table II. The coupling coefficients, k_{33} and k_{31} , of PZT-5H and of BaTiO₃ were directly taken from [12], but the other constants of these three materials were calculated using the data from the same source. The relationship between the measured phase velocities and related elastic constants were derived from the Christoffel wave equations and listed in Table III. Using the four sets of piezoelectric constitutive equations [7], a complete set of elastic, piezoelectric, and dielectric constants of a PZN-

TABLE I
MEASURED PHASE VELOCITIES (M/s) OF ULTRASONIC WAVES IN A PZN-4.5%PT CRYSTAL POLED IN [001].

$v_t^{[001]}$	$v_s^{[001]}$	$v_t^{[100]}$	$v_{s\perp}^{[100]}$	$v_{s\parallel}^{[100]}$	$v_t^{[110]}$	$v_{s\perp}^{[110]}$	$v_{s\parallel}^{[110]}$
4110	2777	3656	2755	2853	4618	668	2855

TABLE II
MEASURED ELECTROMECHANICAL COUPLING COEFFICIENTS AND FREQUENCY CONSTANTS FOR PZN-4.5%PT CRYSTAL POLED IN [001] COMPARED WITH PZT-5H AND BaTiO₃.¹

	$k_{31}^{[110]}$	$2lf_r$	$k_{31}^{[100]}$	$2lf_r$	k_{33}	$2lfa$	k_t	$2lfa$
PZN-4.5PT	0.80	2300	0.50	1210	0.90	2414	0.50	4102
BaTiO ₃ ,crystal ²	0.40	5729	0.32	4542	0.56	3926	0.29	5452
BaTiO ₃ ,ceramic ²	0.21	4391	0.21	4391	0.50	4971	0.43	5477
PZT-5H ²	0.39	2843	0.39	2843	0.75	3851	0.60	4575

¹ Unit for frequency constant = Hertz · meter.
² k_{31} [001] and k_{33} are from [12]. Others are calculated from the material constants in [12].

TABLE III
THE RELATIONSHIPS BETWEEN PHASE VELOCITIES AND ELASTIC CONSTANTS.

	$v_t^{[001]}$	$v_s^{[001]}$	$v_t^{[100]}$	$v_{s\perp}^{[100]}$	$v_{s\parallel}^{[100]}$	$v_t^{[110]}$	$v_{s\perp}^{[110]}$	$v_{s\parallel}^{[110]}$
$\rho v^2 =$	c_{33}^D	c_{44}^E	c_{11}^E	c_{66}^E	c_{44}^D	$\frac{1}{2}(c_{11}^E + c_{12}^E + 2c_{66}^E)$	$\frac{1}{2}(c_{11}^E - c_{12}^E)$	c_{44}^D

TABLE IV
MEASURED AND DERIVED MATERIAL PROPERTIES OF PZN-4.5%PT SINGLE CRYSTAL POLED ALONG [001].¹

Elastic constants: c_{ij} (10^{10} N/m ²)											
c_{11}^{E*}	c_{12}^E	c_{13}^E	c_{33}^E	c_{44}^{E*}	c_{66}^{E*}	c_{11}^D	c_{12}^D	c_{13}^D	c_{33}^{D*}	c_{44}^{D*}	c_{66}^D
11.1	10.2	10.1	10.5	6.4	6.3	11.3	10.4	9.5	13.5	6.7	6.3
±0.15	±0.16	±0.15	±0.3	±0.05	±0.05	±0.15	±0.15	±0.35	±0.5	±0.05	±0.05
Elastic constants: s_{ij} (10^{-12} m ² /N)											
s_{11}^{E*}	s_{12}^E	s_{13}^E	s_{33}^E	s_{44}^E	s_{66}^E	s_{11}^D	s_{12}^D	s_{13}^D	s_{33}^{D*}	s_{44}^D	s_{66}^D
82.0	-28.5	-51.0	108	15.6	15.9	61.5	-49.0	-9.0	20.6	14.9	15.9
±3.2	±0.9	±0.8	±0.5	±0.12	±0.18	±0.8	±0.1	±0.5	±0.8	±0.1	±0.18
Piezoelectric constants: e (C/m ²), d (10^{-12} C/N), g (10^{-3} Vm/N), h (10^8 V/m)											
e_{15}	e_{31}	e_{33}	d_{15}	d_{31}	d_{33}	g_{15}	g_{31}	g_{33}	h_{15}	h_{31}	h_{33}
8.9	-3.7	15.0	140	-970	2000	5.0	-21.0	44	3.4	-4.3	17
±0.9	±1.6	±1.6	±16	±19	±75	±0.58	±0.4	±0.8	±0.4	±2.0	±1.7
Dielectric constants: ϵ (ϵ_0), β ($10^{-4}/\epsilon_0$)								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^*
3000	1000	3100	5200	3.4	10.0	3.2	1.9	0.23	0.50	0.91	0.50
±110	±45	±120	±200	±0.1	±0.5	±0.1	±0.05	±0.02	±0.01	±0.01	±0.01

¹Density: $\rho = 8310$ kg/m³.

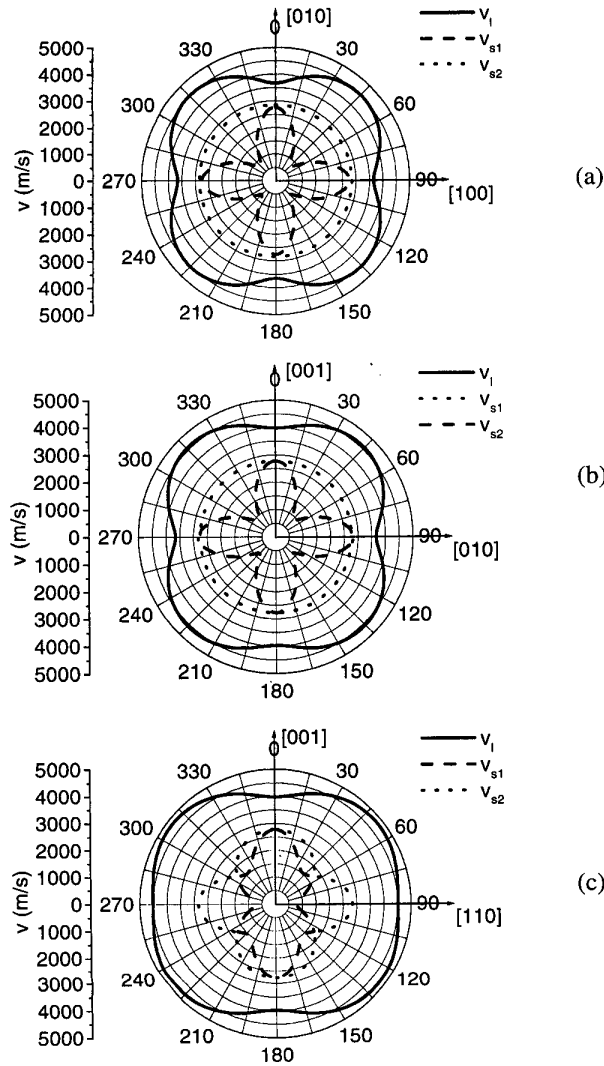


Fig. 2. Directional dependence of the longitudinal velocity, v_l , and the two shear velocities, v_{s1} and v_{s2} in a) the [100] and [010] plane, b) the [010] and [001] plane, and c) the [110] and [001] plane.

4.5%PT crystal based on the 4 mm symmetry has been derived and is given in Table IV. The independent material constants marked with a star (*) in Table IV were measured directly; the other constants in Table IV were derived using the constitutive relations.

Our experimental results confirmed the extraordinarily large room temperature electromechanical coupling coefficient, $k_{33} > 90\%$, and piezoelectric constant, $d_{33} > 2000$ pC/N, reported earlier [3]. More importantly, we have provided a complete set of material constants for this PZN-4.5%PT domain-engineered single crystal, which can be used to perform theoretical analysis and device designs. A very slow shear wave with a sound velocity of only 700 m/s propagating in [110] and polarized in $[\bar{1}\bar{1}0]$ was observed, which implies that a “soft transverse acoustic mode” exists in this domain-engineered single crystal system.

IV. ERROR ANALYSIS

The errors in these measured material constants come from the following sources: 1) the use of parallel plate capacitor approximation for dielectric measurements, 2) errors in measuring the time of flight between ultrasonic echoes, 3) errors in the measurements of resonance and antiresonance frequencies and sample thickness, 4) misorientation and imperfect parallelness of sample surfaces, 5) inconsistency between different samples, and 6) numerical errors for calculating those derived constants. We have analyzed each error source to provide an estimate for the measurement errors.

Using a 15-MHz longitudinal transducer, we have quantified the error for measuring the sound velocity to be $<1\%$ for sample with thickness >2 mm. The error in the capacitance measurement is about 4%. Error from misorientation is $<1\%$ if the misorientation is $<2^\circ$ as analyzed subsequently. The largest error comes from the inconsistency of different samples because the properties of each sample depend strongly on the domain pattern; the domain pattern is controlled by the geometry of the sample and the poling process. This error was minimized through an oversampling scheme in the measurements.

Table V shows the velocities from several measurements in differently orientated samples. One can see that the difference among the measured velocities is $<1\%$. However, because the measured coupling coefficients vary from 1 to 3%, the error of the calculated coefficient $s_{33}^E = s_{33}^D / (1 - k_{33}^2)$ [7] can be very large. This can be seen from the relative error of s_{33}^E caused by the error of k_{33} :

$$\left| \frac{\delta s_{33}^E}{s_{33}^E} \right| = \left| \frac{2k_{33}\delta k_{33}}{(1 - k_{33}^2)} \right|. \quad (1)$$

When k_{33} reaches $>90\%$, the relative error of s_{33}^E calculated from this formula will be an order of magnitude larger than that of k_{33} . For instance, if k_{33} is 0.90 with a relative error of 1%, the relative error for s_{33}^E calculated from (1) will be $>10\%$.

Table VI gives the changes of some constants caused by a 1% change in the coupling coefficient k_{33} . Table VII lists the measured results of the length-extension modes from samples of different aspect ratios. For this system, the aspect ratio (length/width) effects come not only from the influence of mode pureness as stated in [13], but also from the change of the domain patterns. It was found that the coupling coefficient decreases if the aspect ratio decreases, but the frequency constant increases for smaller aspect ratios.

V. ANISOTROPY STUDY

Unlike $\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3$ ceramic samples, domain-engineered PZN-4.5%PT single crystals are anisotropic in the plane perpendicular to the poling direction. Therefore, an analysis of the directional dependence of phase velocities and

TABLE V

MEASURED PHASE VELOCITIES (M/S) OF ULTRASOUND WAVES IN A POLED PZN-4.5%PT CRYSTAL POLED ALONG [001]. THE LEFT COLUMN REPRESENTS THE NUMBER OF MEASUREMENTS. (NOTE: DATA IN THE SAME ROW DOES NOT NECESSARILY MEAN THEY ARE FROM THE SAME SAMPLE.)

Measurements (no.)	$v_l^{[001]}$	$v_s^{[001]}$	$v_l^{[100]}$	$v_{s\perp}^{[100]}$	$v_{s\parallel}^{[100]}$	$v_l^{[110]}$	$v_{s\perp}^{[110]}$	$v_{s\parallel}^{[110]}$
1	4098	2794	3611	2773	2865	4612	649	2864
2	4127	2783	3681	2737	2841	4602	667	2851
3	4122	2766	3639			4612	669	2855
4	4112	2766	3696			4648	688	2842
5	4121		3672					2870
6	4080		3640					2854
7	4123							2860
8								2847
Average	4110	2777	3656	2755	2853	4618	668	2855
Relative error (%)	0.3	0.4	0.7	0.6	0.4	0.3	1.5	0.2

TABLE VI

COMPARISON OF DERIVED CONSTANTS FOR THE TWO CASES OF $k_{33} = 0.90$ AND $k_{33} = 0.91$.

	k_{33}	c_{13}^E	c_{33}^E	c_{13}^D	c_{33}^D	s_{11}^E	s_{33}^E	ϵ_{33}^S	e_{31}	e_{33}	d_{33}	g_{33}	h_{31}	h_{33}
	0.91	9.6	9.5	9.3	13.0	54.1	120	890	-1.5	16.5	2100	46.4	-1.9	20.9
	0.90	10.1	10.5	9.45	13.1	51.4	108	970	-3.7	15.5	2000	43.7	-4.3	17.4
δ^1 (%)	1.1	5.1	10	1.6	0.8	5.1	11	8.6	85	6.3	4.9	6.0	77	18

¹ δ is relative error.

TABLE VII

EXPERIMENTAL RESULTS OBTAINED FROM THE LENGTH EXTENSIONAL RESONANCE USING SAMPLES WITH DIFFERENT ASPECT RATIOS.

l (mm)	l/w	f_r (kHz)	f_a (kHz)	k_{33}	$2lf_a$ (Hz·m)
4.05	9.0	143.9	298.0	0.90	2414
2.90	6.3	222.5	427.5	0.88	2480
3.10	4.5	217.5	408.8	0.87	2534
3.62	3.9	202.4	372.3	0.86	2695

TABLE VIII

RELATIVE ERRORS (IN PERCENTAGES) INTRODUCED IN VELOCITIES WITH A MISORIENTATION OF 2°.

	$v_l^{[001]}$	$v_s^{[001]}$	$v_l^{[100]}$	$v_{s\perp}^{[100]}$	$v_{s\parallel}^{[100]}$	$v_l^{[110]}$	$v_{s\perp}^{[110]}$	$v_{s\parallel}^{[110]}$
Relative error (%)	0.2	0.4	0.3	0.5	0.5	0.06	2.0	0.2

electromechanical coupling coefficients will be very helpful to understand the nature of PZN-PT crystals. The anisotropy analysis was performed by using the measured data given in Table IV. Fig. 2 provides the directional dependence of phase velocities for sound wave propagating in the a) X-Y, b) Y-Z, and c) [110]-Z planes. The calculated results reveal that the velocities of the longitudinal waves do not change with orientation as much as that of the shear waves. As shown in Fig. 2(a), the shear wave propagating in the X-Y plane and polarized in the same plane has the strongest orientational dependence. It has a

maximum in [100] and a minimum in [110], respectively. Fig. 2(b) shows that the phase velocity of the shear wave propagating and polarizing in the Y-Z plane also changes drastically with the propagation direction. Its maximum and minimum are in [001] and [011], respectively. Therefore, the error in phase velocity measurements caused by misorientation is larger for the shear wave propagating in [110] than in other orientations. Table VIII lists the errors of the velocities for an assumed misorientation of 2° in the propagation directions. The maximum error from this error source is 2%.

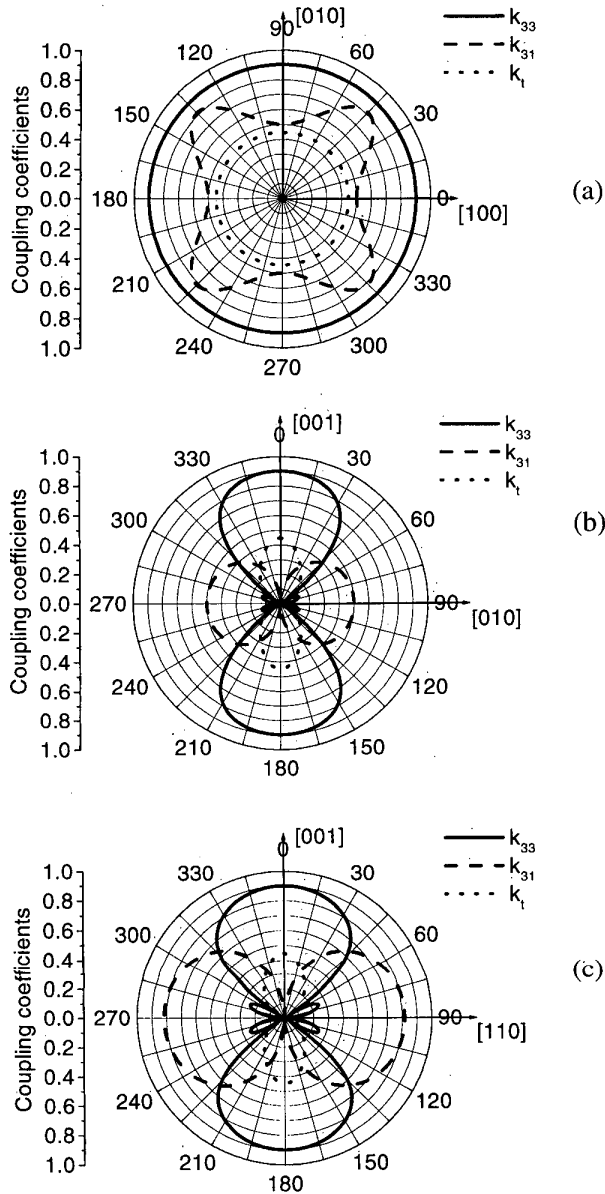


Fig. 3. Directional dependence of the coupling coefficients k_{33} , k_{31} , and k_t , respectively, in a) the [100] and [010] plane, b) the [010] and [001] plane, and c) the [110] and [001] plane.

Fig. 3 shows the directional dependence of the electromechanical coupling coefficients k_{33} and k_{31} . The poling direction is parallel to the length direction for the k_{33} bar and perpendicular to the length direction for the k_{31} bar. The coupling coefficient k_{33} has a maximum value when the length is in [001] [Fig. 3(b)]; the coupling coefficient of k_{31} reaches the maximum when the length is in [110] direction [Fig. 3(c)]. This calculated anisotropy of the lateral coupling coefficient was verified by the direct measurement given in Table II. Fig. 3 also gives the directional dependence of k_t , the coupling coefficient of the thickness mode for a flat plate. It has a maximum in the [001] direction.

Because the 1-D formula for calculating the cou-

pling coefficient and elastic compliance from the length-extensional mode is derived based on the assumption of a long thin bar without lateral constraints, a large aspect ratio is needed to produce such decoupled modes [7]. Otherwise, the measured coupling coefficient from the resonance method will be between the value of k_t and k_{33} . In other words, the longitudinal velocity would be higher for smaller aspect ratios. These results were verified also by the experimental results given in Table VII. It is worth mentioning that some of the modes were not pure even with a very large aspect ratio for this multidomain system; therefore, it is not possible to obtain the complete set of the material properties by using the resonance technique alone.

VI. SUMMARY AND CONCLUSIONS

The material properties of a PZN-4.5%PT single crystal poled in the [001] direction of the original cubic axes were measured using a combined method of ultrasonic and resonance techniques. The crystal symmetry of the ferroelectric state is 3m at room temperature, but the macroscopic average symmetry of the domain pattern is pseudo-tetragonal for the [001] poled samples. Rectangular parallelepiped samples with [001]/[010]/[100] and [001]/[110]/[110] orientations were made for our measurements. A complete set of elastic, piezoelectric, and dielectric constants for the PZN-4.5%PT single crystals were determined from the measured resonance frequencies, sound velocities, and low frequency capacitance values. The directional dependence of the phase velocities of the ultrasonic waves propagating in the X-Y, Y-Z, and [110]-Z planes were calculated based on the measured data. The calculated results showed that the phase velocities are strongly anisotropic for shear waves but directional dependence is relatively weaker for the longitudinal waves. A slow shear wave, about 700 m/s, was found in the [110] direction polarized in [110]. The directional dependence of the electromechanical coupling coefficients was also calculated based on the measured materials properties. It was shown that the maximum value of k_{33} is in [001], and the maximum of effective lateral coupling coefficient k_{31} is in [110]. These calculated results agree well with our experimental verifications. Detailed error analysis was performed for each measured coefficient, and it was concluded that the errors are small for directly measured quantities but relatively larger for the derived coefficients.

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