Characterization of Electromechanical Properties of Relaxor-PT Piezoelectric Single Crystals

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Abstract — In this paper, a new method has been developed to characterize the electromechanical properties of relaxor-PT piezoelectric crystals. In this method, 5 Z-cut samples were used to measure all the eleven electromechanical coefficients of the relaxor-PT piezoelectric crystals by the resonant method. In addition, a technique incorporating laser interferometry was used to precisely measure the piezoelectric constants d_{33} and d_{31}.

INTRODUCTION

Ferroelectric PZN-8% PT crystal belongs to 3m symmetry, with the spontaneous polarization is along the (111) direction. However, the piezoelectric properties of PZN-8% PT crystal are not optimum if the crystal is poled along this direction. It was discovered that PZN-8% PT crystal has extra-high longitudinal electromechanical coupling coefficient k_{33} and piezoelectric constant d_{33} when poled along the (001) direction [1] [2]. For (001) oriented PZN-8% PT crystal, its macroscopic symmetry belongs to 4mm [3].

Although PZN-8% PT crystal exhibits an extra-high k_{33} of approximately 0.94, its thickness mode coupling coefficient k_t is only comparable to PZTs. At higher frequencies (>20 MHz), the monolithic single crystal still offers advantages over PZT in thickness mode operation due to its single grain structure. In order to utilize the advantages of the extra-high k_{33}, 1-3 crystal/polymer composites [4] and transducer arrays incorporating 2-2 composites have been developed. For these two cases the coupling coefficient k_t can approach the k_{33} of the crystal. In order to predict the electromechanical properties of these materials it is necessary to know all eleven electromechanical coefficients of the crystal.

The electromechanical coefficients of crystals can be characterized by IRE Standard [5] only if the specific orientations can be fabricated. The low Curie temperature and ferroelastic nature of PZN-8% PT crystal result in the need for repoling after sample preparation and electroding; therefore, only Z-cut samples can be used.

In this paper, 5 Z-cut samples were prepared and both laser interferometry and resonance methods were used to measure all the eleven electromechanical coefficients of PZN-8% PT crystal. All resonance frequencies were analyzed on an HP 4194 Impedance Analyzer.

EXPERIMENT

For 4mm symmetry, there are six elastic constants, three piezoelectric constants and two dielectric constants.

Piezoelectric constants d_{33} and d_{31}

A sensitive double beam laser interferometer [6] was developed in the Materials Research Lab of The Pennsylvania State University. The system is capable of resolving a displacement of 10^{-2} A° using lock-in detection and measuring the strain all the way to the piezoelectric resonance frequencies using a digital oscilloscope for detection. In this experiment a 1 Volt continuos sinusoidal voltage at 1 kHz was applied to the thickness direction of the crystal sample. The absolute displacements in thickness and width directions were measured using the double beam laser interferometer. The longitudinal strain S_3, lateral strain S_1, and the

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piezoelectric coefficients d_{33} and d_{31} can be calculated as follows:

$$S_3 = \frac{d_3}{t}, \qquad S_1 = \frac{d_1}{w} \tag{1}$$

and

$$d_{33} = \frac{S_3}{E_3} = \frac{d_3}{V},$$

$$d_{31} = -\frac{S_1}{E_3} = -\frac{d_1 t}{Vw} \tag{2}$$

where d_3 and d_1 are the displacements in the thickness and width directions, respectively, and t is the thickness and w is the width of the crystal sample. V and E_3 are the electric voltage and electric field respectively.

Elastic compliance s_{33}^E and coupling factor k_{33}

An (xz) cut thin rod, with a thickness to width ratio of 6, was used to measure the elastic compliance s_{33}^E and coupling factor k_{33} by the resonance method. s_{33}^E and k_{33} can be expressed as follows:

$$k_{33}^2 = \frac{\pi}{2}\frac{f_s}{f_p}\tan\left(\frac{\pi}{2}\frac{f_p - f_s}{f_p}\right) \tag{3}$$

and

$$s_{33}^D = \frac{1}{\rho(2lf_p)^2}$$

$$s_{33}^E = \frac{s_{33}^D}{1 - k_{33}^2} \tag{4}$$

where f_s and f_p are the series and parallel resonant frequencies, respectively, l is the length of the crystal rod, and ρ the density.

Elastic compliance s_{11}^E and coupling coefficient k_{31}

A (zx) cut thin long strip, which satisfies the condition $l_x \gg l_y \gg l_z$, was used to determine the elastic compliance s_{11}^E and coupling factor k_{31}. The formulae used to calculate s_{11}^E and k_{31} are:

$$\frac{k_{31}^2}{1 - k_{31}^2} = \frac{\pi}{2}\frac{f_p}{f_s}\tan\left(\frac{\pi}{2}\frac{f_p - f_s}{f_s}\right) \tag{5}$$

and

$$s_{11}^E = \frac{1}{\rho(2 l_x f_s)^2} \tag{6}$$

where l_x, l_y and l_z are the length, the width and the thickness of the crystal sample respectively.

Elastic compliance $s_{11}^E(45°)$ and coupling factor $k_{31}(45°)$

In order to measure s_{12}^E or s_{66}^E, a (zxt) 45° cut long strip was used to obtain $s_{11}^E(45°)$ and $k_{31}(45°)$ by the resonance method. The formulae are the same as (5) and (6). The relationship among $s_{11}^E(45°)$, s_{12}^E and s_{66}^E is:

$$s_{11}^E(45°) = \frac{1}{2}(s_{11}^E + s_{12}^E) + \frac{1}{4}s_{66}^E \tag{7}$$

where $s_{11}^E(45°)$ is the elastic compliance in the diagonal direction of the x-y plane.

ε_{33}^T, ε_{33}^S, c_{33}^D and k_t

The thickness resonance of a (zx) cut thin square plate resulted in k_t and c_{33}^D. The formulae are:

$$k_t^2 = \frac{\pi}{2}\frac{f_s}{f_p}\tan\left(\frac{\pi}{2}\frac{f_p - f_s}{f_p}\right) \tag{8}$$

and

$$c_{33}^D = \rho(2tf_p)^2$$

$$c_{33}^E = c_{33}^D(1 - k_t^2) \tag{9}$$

where t is the thickness of the plate.

The free dielectric constant ε_{33}^T was calculated from the measured capacitance at 1 kHz. The clamped dielectric constant ε_{33}^S was measured at twice the parallel resonant frequency f_p.

ε_{11}^T, ε_{11}^S, c_{44}^D and k_{15}

The thickness shear resonance of a (yz) cut thin plate determined c_{44}^D and k_{15}. The method is the same as in the thickness mode. The relations are:

$$k_{15}^2 = \frac{\pi}{2}\frac{f_s}{f_p}\tan(\frac{\pi}{2}\frac{f_p - f_s}{f_p}) \qquad (10)$$

and

$$c_{44}^D = \rho(2tf_p)^2$$

$$c_{44}^E = c_{44}^D(1 - k_{15}^2) \qquad (11)$$

$$s_{44}^E = \frac{1}{c_{44}^E}$$

where t is the thickness of the plate. ε_{11}^T and ε_{11}^S are determined in the same way as in thickness mode.

Elastic compliance s_{12}^E, s_{13}^E and s_{66}^E

According to the IEEE Standard, Poisson's ratio σ can be measured by the resonant frequencies of a square thin plate; however, there are several problems with this method. The IEEE Standard assumes that σ is between 0.25 and 0.35. For these crystals it is possible that σ is less than 0.25. Additionally, the IEEE method is correct only for 6mm symmetry, where the ratio of the fourth to the third contour mode frequencies is solely related to σ. For 4mm symmetry this ratio related to both σ and s_{66}^E.

In reality, the resonant modes of a square thin crystal plate are quite complicated. It is difficult to distinguish the origin of the modes. Hence, the IEEE standard cannot be directly used to measure the Poisson's ratio of the PZN-8%PT crystals.

In order to obtain the remaining quantities, the piezoelectric relations of 4mm symmetry were used. The measured quantities k_t, d_{31}, d_{33}, s_{33}^E, s_{11}^E, $s_{11}^E(45°)$, c_{33}^D, ε_{33}^T and ε_{33}^S allowed s_{12}^E, s_{13}^E and s_{66}^E to be calculated according to:

$$e_{33} = k_t\sqrt{c_{33}^D\varepsilon_{33}^S}$$

$$e_{31} = (\varepsilon_{33}^T - \varepsilon_{33}^S - d_{33}e_{33})/2d_{31}$$

$$s_{13}^E = (d_{33} - e_{33}s_{33}^E)/2d_{31} \qquad (12)$$

$$s_{12}^E = s_{11}^E - \frac{2s_{13}^E}{s_{33}^E - 1/c_{33}^E}$$

$$s_{66}^E = 4s_{11}^E(45°) - 2(s_{11}^E + s_{12}^E).$$

RESULTS

The measured electromechanical coefficients for PZN-8%PT are:

$s_{11}^E=72.80\times10^{-12}$ m^2/N,
$s_{12}^E=-13.18\times10^{-12}$ m^2/N,
$s_{13}^E=-57.70\times10^{-12}$ m^2/N,
$s_{33}^E=121.20\times10^{-12}$ m^2/N,
$s_{66}^E=32.75\times10^{-12}$ m^2/N,
$d_{33}=2200\times10^{-12}$ C/N,
$d_{31}=-1075\times10^{-12}$ C/N,
$\varepsilon_{33}^T=5100\varepsilon0$,
$k_{33}=0.94$,
$k_{31}=0.59$,
$k_{31}(30°)=0.74$,
$k_{31}(45°)=0.82$,

$\rho=8.0\times10^3$ kg/m^3.

$c_{11}^E=11.47\times10^{10}$ N/m^2,
$c_{12}^E=10.30\times10^{10}$ N/m^2,
$c_{13}^E=10.37\times10^{10}$ N/m^2,
$c_{33}^E=10.71\times10^{10}$ N/m^2,
$c_{66}^E=3.05\times10^{10}$ N/m^2,
$e_{33}=12.60$ C/m^2,
$e_{31}=-5.83$ C/m^2,
$\varepsilon_{33}^S=560\varepsilon_0$,
$k_t=0.48$.

At this time the electromechanical coefficients related to thickness shear mode are not available.

CONCLUSION

In this paper, the double beam laser interferometer was used to precisely measure the piezoelectric coefficient d_{33} and d_{31}. 5 z-cut samples were used to measure the dielectric and elastic properties of crystal PZN-8%PT by resonance methods.

From the experimental results, it is found that the clamped dielectric constant ε_{33}^S is only $560\varepsilon_0$ although the free dielectric constant ε_{33}^T is $5100\varepsilon_0$.

Since the crystal possesses very large anisotropy, the length of an array element should be along the (100) direction. This orientation is required for achieving the lowest lateral coupling factor k_{31}. The experimental work by Lopath et al. [7] is in agreement with this prediction.

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REFERENCES

[1] J. Kuwata, K. Uchino and S. Nomura, "Dielectric and piezoelectric properties of $0.91Pb(Zn_{1/3}Nb_{2/3})O_3$-$0.09PbTiO_3$ single crystals", Japanese Journal of Applied Physics, Vol. 21, No. 9, pp. 1298, 1982.

[2] S. E. Park and T. R. Shrout, "Characteristics of relaxor-based piezoelectric single crystals for ultrasonic transducers", IEEE Trans. On UFFC, Vol. 44, No. 5, pp. 1140, 1997.

[3] S. Wada, S. E. Park, L. E. Cross and T. R. Shrout, "Domain configuration and Ferroelectric related properties of relaxor based single crystals", Journal of the Korean Physical Society, Vol. 32, pp. S1290, 1998.

[4] T. A. Ritter, K. K. Shung, S. E. Park, X. Geng and T. R. Shrout, "1-3 single crystal composites for ultrasonic transducer arrays", in Proceedings of the SPIE Conference on Ultrasonic Transducer Engineering, pp. 249, 1998.

[5] IRE Standards on Piezoelectric crystals: Determination of the elastic, piezoelectric, and dielectric constants—The electromechanical coupling factor, 1958.

[6] W. Y. Pan and L. E. Cross, "A sensitive double beam laser interferometer for studying high-frequency piezoelectric and electrostrictive strains", Rev. Sci. Instrum., Vol. 60, pp. 2701, 1989.

[7] Lopath et al., "Ultrasonic transducers Using Single Crystal Perovskites", Proc. Intern. Symposium on Applications of Ferroelectrics, pp. 543, 1996.