Influence of sample size on ultrasonic phase velocity measurements in piezoelectric ceramics

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Phase velocities of ultrasound are used for characterizing elastic properties of solids. One of the basic requirements is that the sample area should be much larger than the size of the transducer so that plane wave approximation will hold. This geometric requirement may not be possible to realize for some materials that can only be made in very small size. Using poled and unpoled lead zirconate titanate (PZT-5H) ceramics as examples, we have analyzed experimentally the sample size influence to the ultrasonic measurements. The smallest dimension that is in contact with the transducer is only 14% of the diameter of the transducer. We found that the phase velocity increases when the contact area becomes smaller. The velocity increase is 1.4% and 0.9%, respectively, in the unpoled and poled PZT-5H for the smallest dimension sample compared to bulk values. [DOI: 10.1063/1.1479754]

I. INTRODUCTION

Piezoelectric materials are widely used in various electromechanical devices. Along with density and permittivity, elastic and piezoelectric constants are the fundamental parameters of these materials.

The elastic and piezoelectric constants could be measured by the resonance method based on the IEEE Standards[1] provided samples with different aspect ratios are available and the properties do not have frequency dispersion. Unfortunately, many new materials, particularly some single crystals, are hard to make large enough with uniform properties to satisfy such requirements. The other type of commonly used method for material characterization is the ultrasonic method. This method is based on the measurement of ultrasonic velocities in the material. There are several techniques to measure ultrasonic velocities, for example, pulse-echo, pulse-superposition, and continue wave resonance methods.[2,3] Among these ultrasonic methods, the pulse-echo method is most widely used since it is simple, fast, inexpensive, and reproducible with high accuracy.

It was shown that the hybrid of the resonance and ultrasonic techniques could produce the most self-consistent data set since one could obtain the maximum number of independent measurements from a given sample.[4–11] This hybrid technique has been applied successfully to the newly developed domain engineered PMN-33%PT and PZN-4.5%PT single crystals.[8,9] S. V. Bogdanov *et al.* also developed a characterization technique employing only the ultrasonic waves and requiring fewer samples.[10,11] The question is if the ultrasonic measurement is still reliable enough when the sample dimensions are much smaller than the transducer. The accuracy of the ultrasonic data is very critical for obtaining the complete set of material parameters. We found that 1% error caused by the longitudinal velocity measurement along the [110] direction of PMN-33%PT single crystal could cause the piezoelectric coefficient e_{31} calculation to change more than 30%.

There are several error sources in the ultrasonic wave phase velocity measurements, such as misorientation of crystals, nonparallelness of the sample surfaces, and other system errors.[9] In addition, we have noticed that the phase velocities exhibit strong sample geometry dependence. This problem becomes more pronounced when the sample size is much smaller than that of the transducer. In order to quantify this effect, we have performed a systematic study using poled and unpoled doped lead zirconate titanate (PZT-5H) rectangular rods with the same thickness but different cross section in contact with the transducer. An empirical formula is derived based on the measured data. Self-consistent full set material parameters can be obtained when the data from small size samples are corrected using this correction curve.

II. EXPERIMENTAL SETUP AND SAMPLE PREPARATION

The experimental setup is shown in Fig. 1. One longitudinal transducer with a center frequency of 15 MHz and an element diameter of 0.25 in. was used in the pulse-echo mode. A Panametric 200 MHz computer-controlled pulser/receiver was used to generate a pulse with an energy level of 1 μJ and a damping value of 50 Ω. The wave form from the receiving transducer was recorded by a digital oscilloscope (Tektronix™ TDS 460A) through a 50 Ω coax cable. The sampling rate was 10 Gs/s and the total sampling length for each wave form was 2500 points. The wave form was transferred to a personal computer via a general purpose interface bus for data processing. To reduce random error, each signal was averaged 64 times.

All specimens used in the experiments were made of PZT-5H (Motorola Corp.), and were cut from one large block plate of thickness $H = 5.180 \pm 0.001$ mm. The schematic geometry of the sample and the transducer arrangement are shown in Fig. 2. The dimensions of samples used in the

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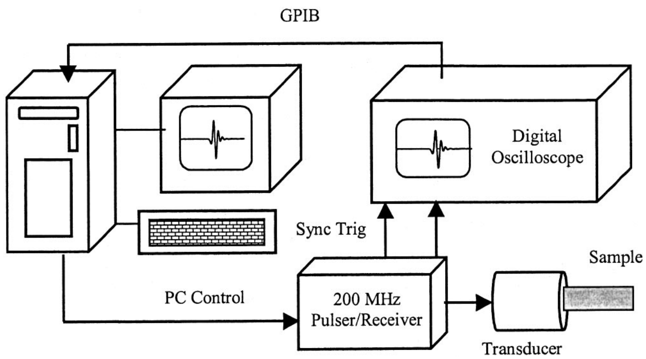

FIG. 1. Experimental setup.

experiments are listed in Table I. The ratio between sample dimension and the diameter of the transducer is also shown in this table. The minimum ratio between the lateral dimension of the sample and the transducer diameter reached 0.14 in our experiments.

III. RESULTS

An ultrasonic pulse-echo method was employed to measure the phase velocity of ultrasonic wave propagating in the PZT-5H samples. As the size of the sample decreases, the first and second echoes distort, making it difficult to judge the corresponding peaks in the echoes. Figure 3(a) shows the signals obtained on samples 3 and 4. One can see that the second echo of the impulse signal from sample 4 has been slightly distorted, which will introduce error to the time-of-flight measurements. In order to resolve this problem, the spectrum valley interval technique was employed in our measurements. The spectrum of the signal enclosing both the first and second echoes was calculated, and the distance between two spectrum valleys near the center frequency of the transducer corresponds to the time of flight between two echoes. As an example, the spectrum of the signal from sample 3 is shown in Fig. 3(b). The relationship between the time of flight of the first and second echoes Δt and the spectrum peak distance Δf is given by

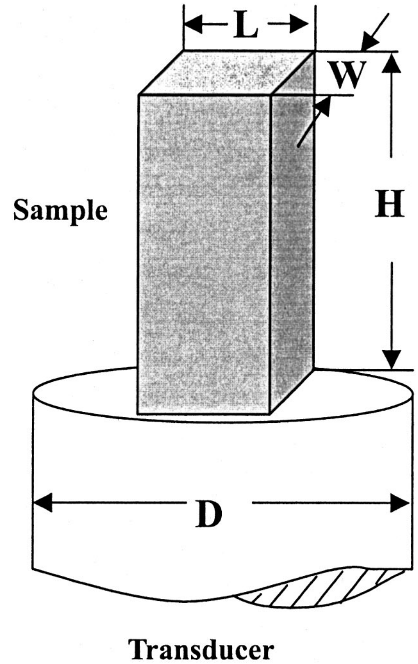

FIG. 2. Schematic geometry of the sample and transducer. The two lateral dimensions L and W were kept the same in our experiments so that the cross section of the sample is maintained as a square.

$$\Delta t=\frac{1}{\Delta f}. \qquad (1)$$

The distance between the two valleys near 15 MHz of the signal spectrum was used to calculate the time of flight of the first and second echoes in this work. Employing this method, the ultrasonic wave phase velocities in the unpoled PZT-5H samples of different sizes were measured, and are shown in Fig. 4. One can see that the ultrasonic wave phase velocity in piezoelectric PZT-5H ceramic rods increased about 1.4% when the lateral dimension of the sample reduces to 14% of the diameter of the transducer. The ratios of lateral dimension to the ultrasonic wavelength are also listed in Table I, in which λ_U is the ultrasonic wavelength in unpoled PZT-5H samples. A smaller sample with dimension less than

TABLE I. The dimensions of the PZT-5H samples used in our experiments.

Sample No.	L (mm)	W (mm)	H (mm)	L/D	L/λ_U	L/λ_P
1	25.40	25.40	5.18	4.0000	90.6170	81.7318
2	12.70	12.70	5.18	2.0000	45.2995	40.8661
3	6.69	6.69	5.18	1.0535	23.8253	21.4972
4	5.85	5.85	5.18	0.9213	20.8229	18.7904
5	4.90	4.90	5.18	0.7717	17.4250	15.7278
6	3.10	3.10	5.18	0.4882	10.9903	9.9291
7	2.60	2.60	5.18	0.4094	9.2048	8.3198
8	2.46	2.46	5.18	0.3874	8.7068	7.8708
9	2.26	2.26	5.18	0.3559	7.9940	7.2281
10	1.97	1.97	5.18	0.3102	6.9609	6.2964
11	1.60	1.60	5.18	0.2520	5.6472	5.1109
12	1.48	1.48	5.18	0.2331	5.2211	4.7263
13	1.22	1.22	5.18	0.1921	4.2979	3.8925
14	0.93	0.93	5.18	0.1465	3.2731	2.9662
15	0.87	0.87	5.18	0.1370	3.0599	2.7734

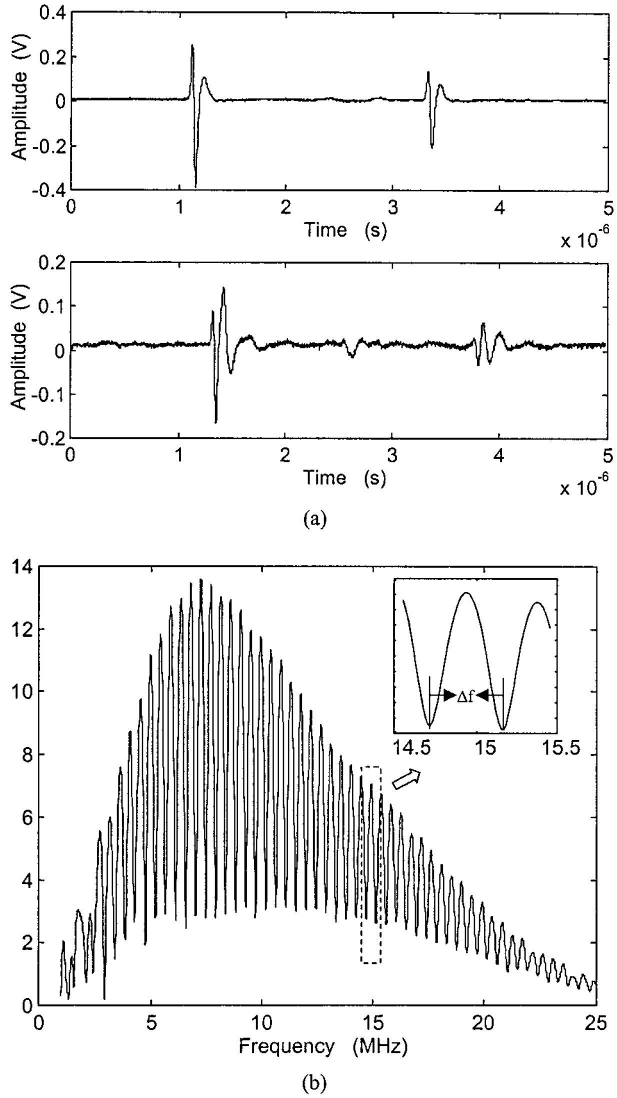

FIG. 3. (a) The time domain signal of the first and second ultrasonic echoes in sample 3 and 4, respectively; (b) the spectrum of signal from sample 4.

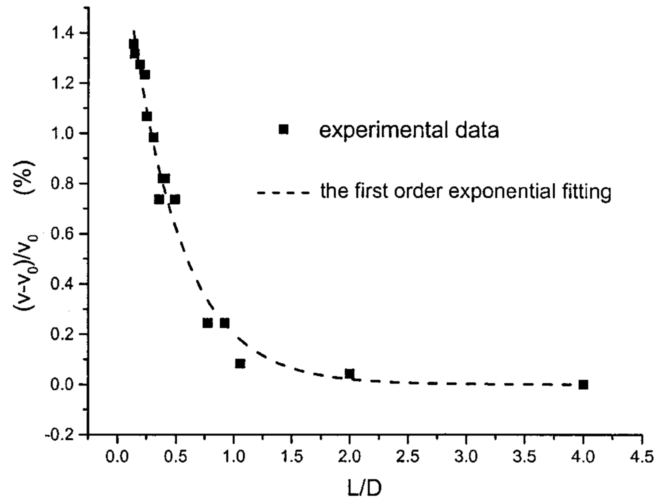

FIG. 4. Ultrasonic wave phase velocities in unpoled PZT-5H samples of different lateral dimensions.

$3\lambda_U$ was tested, but the second echo could no longer be recognized under the signal-to-noise ratio condition of our measurement system. It can be seen from Table I that the minimum ratio of sample lateral dimension to the ultrasonic wavelength is 3.0.

Based on the shape of the curve in Fig. 4, we have fitted the data to an exponential form so that the relationship between the phase velocity in unpoled PZT-5H ceramic and the ratio $r = L/D = W/D$ is given by:

$$\nu = 4204.5*(1+0.01916e^{-r/0.4429}). \tag{2}$$

Poled PZT-5H samples were also measured in this work. All samples were poled under the electric field of 10 kV/cm for 3 min. The ultrasonic phase velocity measurement was repeated on each sample. The relationship between the phase velocity of the poled PZT-5H samples and the sample to transducer ratio r is shown in Fig. 5, in which the piezoelectric constants d_{33} of all samples were also measured and plotted. From Fig. 5, the same trend of phase velocity versus sample dimension could be seen, except two points generated from samples with the ratio $r = L/D$ around 0.35 and 0.13. We found that most of the d_{33} values of these samples are around 690 ± 5 pC/N, but d_{33} of these two samples are 676 and 683 PC/N, respectively, which means that they are underpoled.

In order to verify this point, the relationship between d_{33} and the phase velocity on the same sample was measured and the result are shown in Fig. 6. All the measurements were performed on sample 1 which was depoled and repoled under different poling condition to obtain different d_{33} values. Since the piezoelectric constant e_{33} and the dielectric constant ε_{33} will all change under different poling condition, the elastic constant $c_{33}^D = c_{33}^E + e_{33}^2/\varepsilon_{33}^S$ will be affected, producing a change to the phase velocity $\nu = \sqrt{c_{33}^D/\rho}$. It can be seen that the ultrasonic phase velocity in PZT-5H sample increases nonlinearly with d_{33}. Based on the results in Fig. 6, we have used a second order polynomial to interpret the data:

$$\nu = 4195.8 + 0.32885*10^{12}*d_{33} + 5.97*10^{20}*d_{33}^2. \tag{3}$$

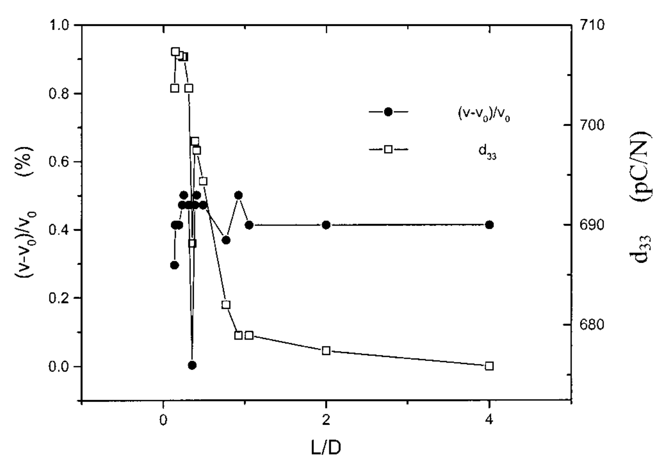

FIG. 5. Ultrasonic wave phase velocities and their d_{33} in poled PZT-5H samples of different lateral dimensions.

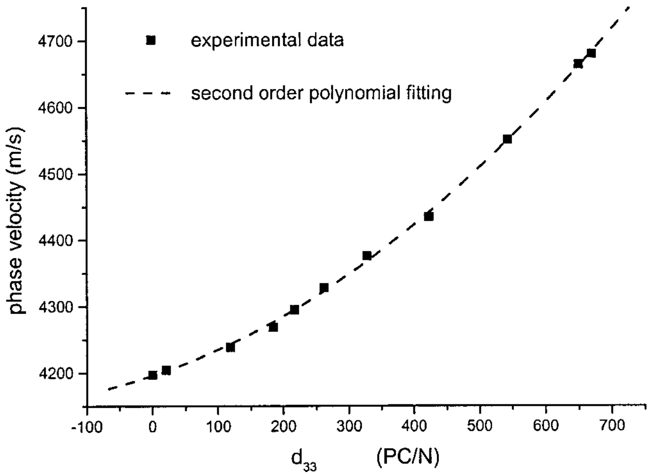

FIG. 6. The piezoelectric coefficient d_{33} vs ultrasonic wave phase velocity measured on sample 1.

Using Eq. (3), the values in Fig. 5 were corrected so that the d_{33} of all samples were calculated based on the same $d_{33} = 690$ pC/N. The corrected data are plotted in Fig. 7.

It is shown in Fig. 7 that the longitudinal wave velocity increases about 0.9% when the lateral dimensions of the sample become about 14% of the diameter of the transducer. The ratios of the sample lateral dimension to the ultrasonic wavelength are also listed in Table I, in which λ_P is the longitudinal ultrasonic wavelength in poled PZT-5H samples. An empirical equation for the relationship between the phase velocity of poled PZT-5H ceramic and L/D ratio r was obtained by fitting the data:

$$\nu = 4662.5*(1+0.01352e^{-r/0.4699}). \tag{4}$$

All ultrasonic measurement methods are under the assumption that the ultrasonic wave is a perfect plane wave. This assumption holds true only when the wavelength is much smaller than the sample size and the size of sample is much larger than that of the transducer. In reality, samples having size larger than the transducer is not always possible, we have encountered this problem in the newly developed PMN-PT, PZN-PT, and PYN-PT single crystals, which have some composition nonuniformity for larger samples and the properties are extremely sensitive to the composition. The assumption of plane wave could no longer be satisfied as the size of the sample became comparable to the wavelength and the size of the sample becomes much smaller than the transducer. Under such circumstances, the waves start to behave like guided waves rather than a pure longitudinal plane wave. This guided wave is different from the Lamb wave in plate with free boundary or Rayleigh waves on the boundary between a half-space and vacuum because both the length and width of the ceramic samples are comparable to the wavelength.[12] Similar problems had been studied by J. L. Rose *et al.* for an ultrasonic wave propagating in a cylindrical rod with the rod diameter comparable to or less than the ultrasonic wavelength, the wave propagation direction is along the axis of the cylindrical rod.[13] They found that there could be more than one mode propagating along the rod when the diameter becomes comparable to the wavelength, in which the fundamental mode of the rod is a kind of longitudinal mode with the displacement of the particle parallel to the direction of propagation, and the phase velocity nonlinearly increases as the diameter of the cylinder decreases. Although the modes being excited in the rectangular rods might be different from those in a cylindrical rod, the fundamental mode is similar. Our experimental results have verified this point, i.e., the phase velocity shows increase with decreasing the lateral dimension of the sample.

Our empirical formula can be used to adjust the experimental value of ultrasonic phase velocity obtained from smaller samples to reflect the true bulk value. Such results are particularly useful when both large and small samples are needed in determining a complete material data set of a piezoelectric material. It can greatly enhance the self-consistency of full matrix material property data derived from measurements using both large and small size samples.

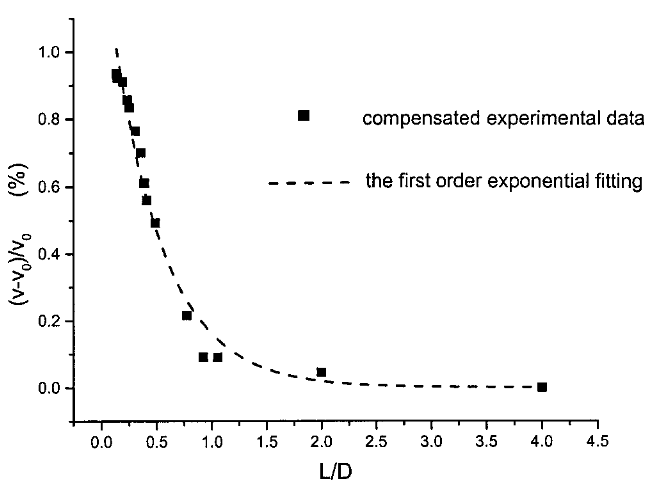

FIG. 7. Compensated ultrasonic wave phase velocity in the poled PZT-5H samples of different lateral dimensions.

IV. DISCUSSION AND CONCLUSIONS

The ultrasonic wave velocity propagating in the unpoled and poled PZT-5H ceramics have been measured using different size samples. It has been observed that the longitudinal velocity increased 1.4% and 0.9%, respectively, for poled and unpoled PZT-5H samples when their lateral dimensions decrease to 14% of the transducer diameter. An exponential empirical relationship between the sample to transducer ratio and the wave velocity has been fitted, which provides a useful correction standard for the accurate determination of material constants of bulk PZT when the sample size is small.

It is worth mentioning that the resonance ultrasonic spectroscopy (RUS) technique is another powerful method developed in recent years for the characterization of elastic properties,[14,15] which is especially suitable for small samples. Through analysis of the resonant ultrasound spectrum of a small rectangular parallelepiped sample, the complete set of elastic constants of this material can be derived using a regression technique. The technique can be also used to characterize piezoelectric materials if the mechanical Q is high enough.[16] Compared with conventional pulse-echo method, the RUS technique needs only one sample, hence producing more self-consistent data. On the other hand, the data analysis becomes very uncertain for lower symmetry crystals as the number of unknowns becomes large. Most piezoelectric materials used today are ceramics and their Q values are not very high. This makes the RUS method difficult to apply since there are not enough resonance peaks to uniquely determine all the coefficients at the same time. In addition, as the number of unknown constants increases, the inversion procedure becomes less unique since the minimization process depends strongly on the initial guess, and there are multiple solutions. On the other hand, the strategy presented in our paper provides a simpler and more direct way to deal with small samples. The results are unambiguous.

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