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Temperature Dependence of Sound Velocity in Tin Modified Lead Zirconate Titanate Ceramics

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Abstract

Sound velocity traveling through unpoled and poled tin modified lead zirconate titanate ceramics (PSZT) was measured by a pulse-echo/through transmission buffer rod technique from sub-ambient to above its Curie temperatures. Elastic moduli were determined from elastic constants that were calculated from acoustic velocities assuming a transverse isotropic and a cylindrical symmetry for unpoled and poled samples respectively. Results indicated that both longitudinal and shear velocities changed significantly near phase transformation temperatures. In addition all elastic moduli including Young's modulus, shear modulus, and bulk modulus reached a local minimum at phase transformation temperatures with a global minimum at its Curie temperature. Details of the temperature dependence of acoustic measurements as well as the change in elastic behavior for the PSZT ceramics will be presented.

Material – PSZT Ceramics

Lead zirconate ($PbZrO_3$ or PZ) and lead titanate ($PbTiO_3$, PT) form a continuous series of solid solutions over the entire composition range. The solid solutions, commonly referred as PZTs or $Pb(Zr_xTi_{1-x})O_3$, contain a variety of ferroelectric (FE) and non-ferroelectric phases and are the backbones for many ferroelectric and piezoelectric applications. The specific composition for this study is a tin modified PZT (or PSZT) which locates on the zirconia-rich side of the PZT phase diagram near the boundary between FE and anti-ferroelectric (AFE) phases. Above its Curie temperature the material possesses an ideal cubic perovskite (*Pm3m*) structure. Upon cooling, the paraelectric (PE) phase first transforms into a high-temperature rhombohedral R3m (FE_{RH}) phase and later transforms into a low temperature rhombohedral R3c (FE_{RL}) phase above room temperature. All of these phase transformations are known to be associated with lattice instability or soft phonon modes which provide many interesting property changes for practical applications.

Poled and Unpoled Ferroelectric Ceramics

One of the changes accompanied with structural phase transformations is the development of a random domain structure during the PE-to-FE phase transformation. These random domains, each with a unique macroscopic polarization direction, can be aligned by an external field through a poling process and render piezoelectric responses to ferroelectric ceramics. The alignment of domains along the poling direction also introduces a polar cylindrical symmetry and anisotropic responses to the originally unpoled isotropic ceramics. This work will study the changes of acoustic and elastic properties before and after the poling process.

Temperature Dependence of Acoustic Measurements

In this work, the sound velocity traveling through poled and unpoled ceramics was measured by a pulse-echo/through transmission buffer rod technique from -65°C to 250°C. The experimental technique involves propagation of a series of acoustic waves through two buffer rods with a sample between them. Figure 1 shows a simple setup of the experiment (without showing the cold and hot stages). The time of flight in conjunction with the thickness of the sample can be used to estimate the acoustic velocity through the material of interest. Based on this simple setup, the time through the sample (t_S) can be expressed by the transit time through the entire system (t_T), and the transit time through each buffer rod (t_{PE1} and t_{PE2}).

$$t_S = t_T - \frac{1}{2}(t_{PE1}+t_{PE2})$$

Elastic modulus can be calculated by the following equations:

- $\sigma = \frac{1-2(v_t/v_l)^2}{2-2(v_t/v_l)^2}$
- $E = \frac{v_l^2\rho(1+\sigma)(1-2\sigma)}{(1-\sigma)}$
- $G = v_t^2\rho$
- $B = \frac{EG}{3(3G-E)}$

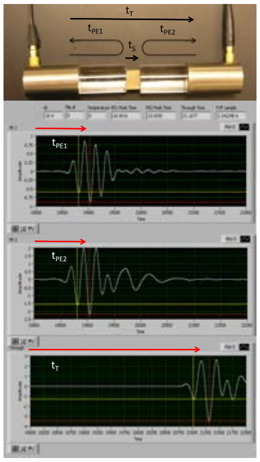
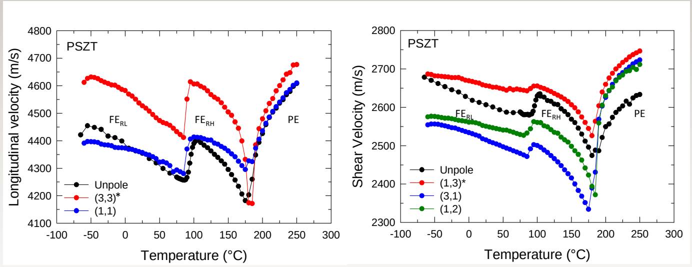


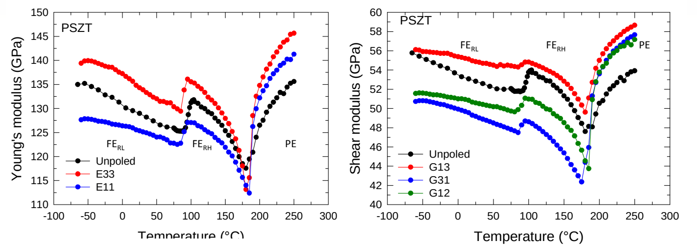
Fig. 1 Acoustic measurement data analysis.

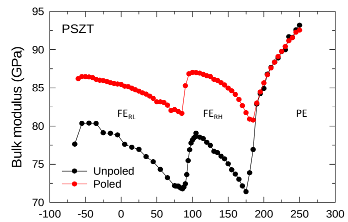
Acoustic Velocity



- Both longitudinal and shear velocities reach local minima at phase transformation temperatures.
- Piezoelectric stiffening increases the sound speed in (3,3) and (1,3) directions.

Elastic Modulus





- Anisotropic behavior is observed after the poling process.
- All elastic modulus reach local minima at phase transformation temperatures.
- The global minimum of elastic moduli can be found at the PE-to-FE phase transformation temperature.

Summary

- Dielectric stiffening creates anistropic behavior in poled PSZT ceramics.
- Acoustic velocities and elastic moduli reach local minima at phase transformation temperatures.
- The changes of acoustic velocities and elastic moduli at phase transformation temperatures are consistent with soft phonon behavior.

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