Shear piezoelectric coefficients of PZT, LiNbO$_3$ and PMN-PT at cryogenic temperatures

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Abstract.
Piezoelectric transducers are used to detect stress and to generate nanometer scale displacements but their piezoelectric coefficients decrease with temperature, limiting their performance in cryogenic applications. We have developed a capacitive technique and directly measured the temperature dependence of the shear coefficient $d_{15}$ for ceramic lead zirconium titanate (PZT), 41° X-cut lithium niobate (LiNbO$_3$) and single crystal lead magnesium niobium-lead titanate (PMN-PT). In PZT, $d_{15}$ decreases nearly linearly with temperature, dropping by factor of about 4 by 1.3 K. LiNbO$_3$ has the smallest room temperature $d_{15}$, but its value decreased by only 6% at the lowest temperatures. PMN-PT had the largest value of $d_{15}$ at room temperature ($2.9 \times 10^{-9}$ m/V, about 45 times larger than for LiNbO$_3$) but it decreased rapidly below 75 K; at 1.3 K, $d_{15}$ was only about 8% of its room temperature value.

1. Introduction

A mechanical stress applied to a piezoelectric material produces a charge proportional to the force; a voltage applied across a piezoelectric produces a strain. These properties can be exploited to measure stresses and to generate displacements in mechanical actuators. Although most applications are at room temperature, scanning tunneling microscopes (STM) are often designed to operate below 4 K, and we have used PZT transducers to measure the elastic and plastic properties of solid helium [1, 2] at temperatures as low as 15 mK. For high sensitivity and/or maximum displacements, large piezoelectric coefficients $d_{ij}$ are needed, but most materials’ piezoelectric coefficients are reduced at cryogenic temperatures. The most commonly used material, PZT, has relatively large piezoelectric coefficients at room temperature. However, $d_{33}$ and $d_{13}$ are about 5 times smaller at 4 K [3, 4, 5, 6] and little is known about the shear coefficient $d_{15}$ at cryogenic temperatures. The temperature dependence for lithium niobate is much weaker, but its largest coefficient, $d_{15}$, is about 8 times smaller than for PZT [7, 8]. New materials like single crystal PMN-PT have exceptionally large room temperature coefficients (e.g. $d_{15}$ about
5 times larger than for PZT) but limited information is available about their behavior at low temperatures, particularly below 75 K [9, 10]. Also, the displacements in piezoelectrics often have non-linear or time-dependent behavior [12, 11, 13] which produces hysteresis when large voltages are applied and limits their performance in STM or other positioning applications.

In this paper we report measurements of $d_{15}$ for ceramic PZT 5A[14], $41^\circ$ X-cut LiNbO$_3$[15] and single crystal PMN-PT[16] transducers. Measurements were made between room temperature and 1.3 K, at voltages up to 150 V. The large room temperature values of $d_{15}$ for PMN-PT are reduced by a factor of 12 at the lowest temperatures, compared to a 4-fold reduction for PZT. At the lowest temperatures, all three materials show linear, non-hysteretic behavior up to 150 V, but the PMN-PT transducers have significant hysteresis above 100 K.

2. Experimental design

In order to directly measure the shear piezoelectric coefficient $d_{15}$ at low temperature, we used a capacitive displacement detection technique as shown in Fig. 1. A voltage $\Delta V$ applied across the shear transducer induces a transverse displacement $\Delta x = d_{15} \Delta V$ which then produces a capacitance change $\Delta C$ proportional to the change in spacing $\Delta L = \Delta x$ between the two blocks

$$\frac{\Delta C}{C} = \frac{\Delta L}{L} = \frac{d_{15} \Delta V}{L},$$

(1)

where the gap spacing $L$ (typically about 300 $\mu$m) is related to the initial capacitance $C = \epsilon_0 A / L$, where $A$ is the area of the gap where the blocks overlap. The piezoelectric coefficient is then

$$d_{15} = A \epsilon_0 \frac{\Delta C}{C^2} \frac{\Delta V}{\Delta V}.$$  

(2)

The capacitance/transducer assembly was mounted on a copper bracket attached to the $^4$He pot of a dilution refrigerator. Above 4 K, helium exchange gas was used for cooling.

![Figure 1. Displacement measurement to determine $d_{15}$. A Keithley 2400 voltage source applies an alternating $\pm 20$ V potential to the transducer’s electrodes. Coaxial cables connect a capacitance bridge[17] to a fixed brass block and to a block clamped on top of the shear transducer. A displacement of the transducer changes the gap between the two blocks and their capacitance.](image-url)
Figure 2. Capacitance changes due to voltage ±20V applied to a PZT-5A transducer at 20 K.

Figure 2 shows typical capacitance changes when an alternating DC voltage ±20 V ($\Delta V = 40$ V) is applied across the PZT-5A shear transducer at 20 K. The average capacitance $C = 14.0985$ pF and the change $\Delta C$ (about 56 ppm) give $d_{15} \approx 1.8 \times 10^{-10}$ m/V.

3. Results

The temperature dependences of the shear piezoelectric constant for the ceramic PZT-5A and 41° X-cut LiNbO$_3$ shear transducers are plotted in Fig. 3. The error bars for LiNbO$_3$ correspond to the resolution of our capacitance bridge; the errors for PZT and PMN-PT are smaller than the symbol sizes. For PZT (Fig. 3a), $d_{15}$ decreases roughly linearly over the entire temperature range. Before taking data, the temperature was stabilized at each point for at least 50 minutes; a complete cooling or warming run took at least 4 days. The data were during cooling (green points) and during warming (red points) agree. Thermally cycling to low temperature did not affect the room temperature properties of the PZT or other transducers. The measured value of $d_{15}$ for our PZT transducer decreased from $6.1 \times 10^{-10}$ m/V at room temperature (close to the value quoted by the manufacturer: $5.85 \times 10^{-10}$ m/V). At our lowest temperature (1.3 K), this dropped to $1.44 \times 10^{-10}$ m/V, a reduction of more than a factor of 4. Figure 3b shows the corresponding data for LiNbO$_3$. The room temperature value of $d_{15}$ is about ten times smaller, $6.4 \times 10^{-11}$ m/V, but only drops by about 6% at low temperature.

The corresponding temperature dependence for the PMN-PT single crystal is shown in Fig. 4a. At room temperature $d_{15}$ has a value of $2.85 \times 10^{-9}$ m/V in this material, about 5 times larger than PZT. It decreases with temperature, but is nearly constant at $2.0 \times 10^{-9}$ m/V between 225 and 75 K. However, below 75 K, $d_{15}$ drops rapidly, by a factor of nearly 10 at 1.3 K.

4. Discussion

At room temperature, $d_{15}$ of the three materials varies by a factor of 45, from $6.4 \times 10^{-11}$ for LiNbO$_3$ to $2.85 \times 10^{-9}$ m/V for PMN-PT. In all of the materials, $d_{15}$ decreases with temperature, but the variation is smallest for LiNbO$_3$ and largest for PMN-PT. At 1.3 K, $d_{15}$ ranges from
Figure 3. a) Temperature dependence of the piezoelectric coefficient $d_{15}$ of a PZT-5A shear transducer. Green diamonds are data taken during cooling; red circular points during warming. b) Temperature dependence of $d_{15}$ for a 41° X-cut LiNbO$_3$ crystal.

Figure 4. a) The temperature dependence of $d_{15}$ for a PMN-PT single crystal shear transducer. b) Comparison of the shear coefficients of PZT, LiNbO$_3$ and PMN-PT transducers.

$6.0 \times 10^{-11}$ for LiNbO$_3$ to $2.3 \times 10^{-10}$ m/V for PMN-PT, less than a factor of 4. Single crystal PMN-PT does have a significant advantage over more commonly used PZT ceramics at intermediate temperatures. At 75 K its value of $d_{15}$ is about 8 times larger than that of PZT-5A but this advantage nearly disappears at lower temperatures (at 1.3 K it is only 60% larger). Lithium niobate has the smallest $d_{15}$ at all temperatures, but at 1.3 K it is nearly half the value for PZT-5A.

Lithium niobate has a very high ferroelectric Curie temperature (1415 K) and its relatively small piezoelectric coefficients and weak temperature dependence reflect the intrinsic behavior of single domain crystals. The much larger room temperature coefficients of PZT-5A ceramics and PMN-PT crystals are achieved with compositions close to a transition between tetragonal and rhombohedral structures. Their piezoelectric behavior is determined by the temperature dependence of this phase boundary and by extrinsic effects associated with grain boundaries and domain walls, which we plan to study in future work.
In addition to the measurements described above, we also measured the displacements when voltages between -150 and +150 V were applied. Over this voltage range, the response was linear at all temperatures for LiNbO$_3$ and PZT. At the lowest temperatures this was also true for PMN-PT but the response was non-linear in the plateau region above 75 K. This resulted in hysteresis in the displacement when the voltage was ramped up and down between -150 and +150 V, limiting the use of PMT-PT in applications like STMs where precise, reproducible positioning is required.

Lithium niobate shear transducers have some advantages for low temperature actuators. In addition to the linear response described above, LiNbO$_3$ shear transducers can tolerate much higher voltages than PZT or PMN-PT without depoling. Also, they can be cut much thinner than PZT or PMN-PT shear transducers, which are usually at least a millimeter thick to allow them to be electrically poled parallel to the transducer surface. A compact, large displacement, linear actuator could be constructed from a stack of thin LiNbO$_3$ transducers.

In other applications, e.g. precision stress measurements, the calibration depends on $d_{15}$ and therefore depends on temperature. For LiNbO$_3$, $d_{15}$ is essentially constant (within the resolution of our measurements) below 100 K. For PZT and PMN-PT, on the other hand, $d_{15}$ continues to change to the lowest temperatures, dropping by about 4% and 6%, respectively, between 4 and 1.3 K.

4.1. Acknowledgments

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[15] 41° X-cut lithium niobate shear transducers, 10.0 × 10.0 × 0.25 mm with chrome/gold electrodes, supplied by Boston Piezo-Optics, Inc.
[16] TRS X2A single crystal lead magnesium niobate-lead titanate (PMN-PT) shear plates, 10.0 × 10.0 × 1.0 mm with chrome/gold electrodes, supplied by TRS Ceramics, Inc. (http://www.trstechnologies.com/)
[17] Andeen-Hagerling 2550A 1 kHz AC capacitance bridge with a resolution of about 0.5 ppm for a typical averaging time of 9 seconds (http://www.andeen-hagerling.com/).