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Young's modulus and hysteretic losses of $0.7Pb(Mg_{1/3}Nb_{2/3})O_3-0.3PbTiO_3$: single versus polycrystalline forms

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Young's modulus (Y) of $0.7Pb(Mg_{1/3}Nb_{2/3})O_3-0.3PbTiO_3$ has been investigated for polycrystals and single crystals oriented along ⟨001⟩, ⟨110⟩, and ⟨111⟩. The value of $Y_{\langle 001\rangle}$ and $Y_{\langle 110\rangle}$ for single crystals was dramatically lower than either $Y_{\langle 111\rangle}$ or the polycrystalline averaged value. For ceramics, field dependent measurements revealed a significant softening of Y, which was not observed for oriented crystals. For both forms, the elastic energy densities were similar, however significantly higher hysteretic losses were found for polycrystals. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1618940]

INTRODUCTION

Single crystals of $0.7Pb(Mg_{1/3}Nb_{2/3})O_3-0.3PbTiO_3$ (PMN–PT) that are oriented along the ⟨001⟩ direction have high electromechanical coupling coefficients ($k\sim 0.95$) and electrically induced strains.[1–3] These properties are superior to those of $Pb(Zr_{1-x}Ti_x)O_3$ (PZT) ceramics that have traditionally been used in transducers and actuators.[4] ⟨001⟩-oriented single crystals of $0.7Pb(Mg_{1/3}Nb_{2/3})O_3-0.3PbTiO_3$ are currently under development for advanced transducers in sonar. In acoustic transducer applications,[5] uniaxial prestress inherently needs to be used. The influence of applied uniaxial stress σ on the electromechanical performance characteristics of ⟨001⟩-oriented PMN–PT crystals has recently been investigated.[6,7] These investigations have demonstrated a depolarization of the crystals with increasing σ. The electromechanical coupling and acoustic power density are relatively σ independent over a modest range of stresses, because the elastic strain and polarization are dually changed under σ.

It has been known for some time that poled PMN–PT ceramics and soft PZTs have high coupling and piezoelectric constants,[8–10] however, their application in acoustics and transduction has been limited by hysteretic losses, resulting in thermal stability problems. Even though hard PZT materials have lower electromechanical performance coefficients,[8] acoustic and transduction devices are designed around these compositions, in order to reduce the hysteretic losses. The performance characteristics of ⟨001⟩-oriented $0.7Pb(Mg_{1/3}Nb_{2/3})O_3-0.3PbTiO_3$ crystals are significantly superior to those of corresponding compositions of PMN–PT ceramics.[1–3,9,10]

The purpose of this work was to investigate the elastic and hysteretic properties of poly- and single crystal forms of the same PMN–PT composition as a function of σ. Both forms of the material have been found to have similar electroacoustic energy densities, however the ceramic form has significantly higher hysteretic losses.

Single crystals of PMN–PT 70/30 that are oriented along the ⟨001⟩, ⟨110⟩, and ⟨111⟩ directions were obtained from HC Materials (Urbana, IL). These crystals were grown by a seeded vertical Bridgman method.[11–13] Corresponding PMN–PT 70/30 ceramics were fabricated using conventional mixed oxides methods. These specimens were prepared by EDO Corporation (Salt Lake City, UT). $\epsilon-\sigma$ measurements were performed on bar shaped specimens having an aspect ratio of 4:1, which had a length of ~1 cm. Strain gauges were mounted on the side of the specimen and a mechanical load was applied using a pneumatic cylinder. Special care was taken to ensure a uniform stress distribution in the setup by: (i) placing the strain gauges in the center of the specimen, where the stress is constant; (ii) using rectangular shaped specimens, with parallel sides; and (iii) application of a uniform load on the specimen, via the pneumatic cylinder. $\epsilon-\sigma$ measurements were performed under zero dc bias and at various dc bias levels (applied along the load axis). Young's modulus (Y) was determined from the slope of the $\epsilon-\sigma$ curve. In addition, $P-E$ and $\epsilon-E$ measurements were simultaneously performed using a modified Sawyer–Tower bridge. The area of the hysteresis loop of the $P-E$ curve was determined by integration using Green's theorem.

Figure 1(a) shows the $P-E$ curve for the polycrystalline form taken under unipolar drive at various σ. The induced polarization at 15 kV/cm, $\Delta P_{(15\ kV/cm)}$, can be seen to be increased from ~0.06 to 0.16 C/m^2 with increasing uniaxial stress between 0 and 6×10^7 N/m^2. Figure 1(b) shows the unipolar $P-E$ response for a ⟨001⟩-oriented crystal under various σ. This figure reveals a significant increase in $\Delta P_{(15\ kV/cm)}$ relative to the ceramic form. $\Delta P_{(15\ kV/cm)}$ increased from ~0.08 C/m^2 under small loads to ~0.25 C/m^2 under 6×10^7 N/m^2. The data in the figures show that the value of the remanent polarization P_r under zero E shifts with σ, due to a partial depoling with increasing σ. The data also reveal an increase in the hysteretic losses with increasing σ. The $\epsilon-E$ data are shown at various mechanical stresses in Figs. 1(c) and 1(d) for the polycrystalline and ⟨001⟩-oriented crystal, respectively. These data exhibit the same general trends of increasing ϵ with σ. However, the value of ϵ under 15 kV/cm, $\epsilon_{(15\ kV/cm)}$, was significantly larger for the single crystal relative to polycrystal form.

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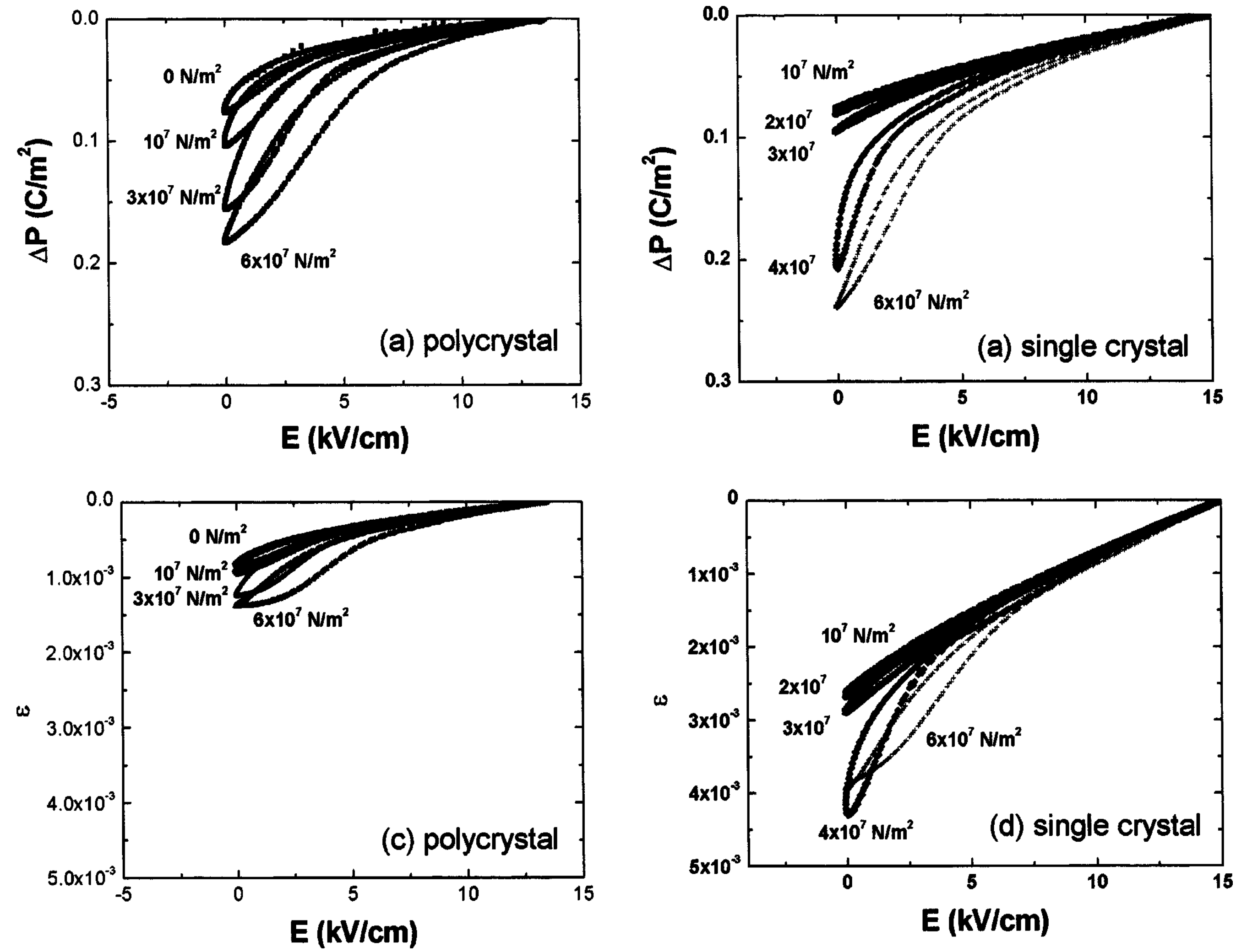

FIG. 1. $P-E$ and $\epsilon-E$ responses at different uniaxial stresses between 0 and 6×10^7 N/m^2 under unipolar drive conditions: (a) $P-E$ response for a ceramic specimen, (b) $P-E$ response for ⟨001⟩-oriented crystal, (c) $\epsilon-E$ response for a ceramic specimen, and (d) $\epsilon-E$ response for ⟨001⟩-oriented crystal.

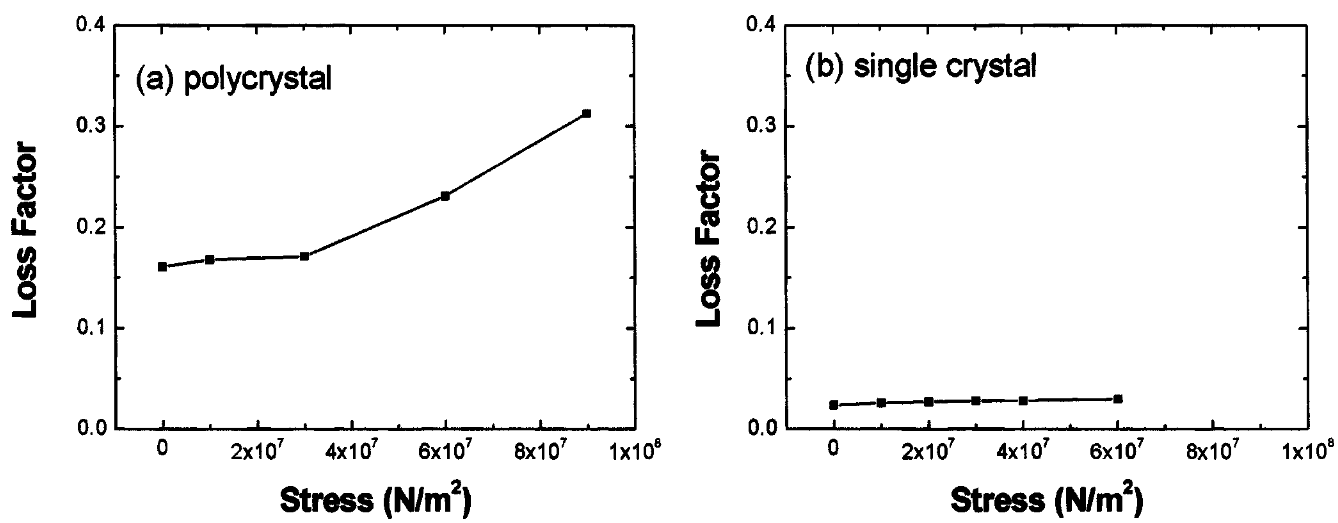

FIG. 2. Hysteretic loss as a function of σ for single and polycrystalline PMN–PT 70/30 for $E=15$ kV/cm: (a) polycrystalline form and (b) single crystal form.

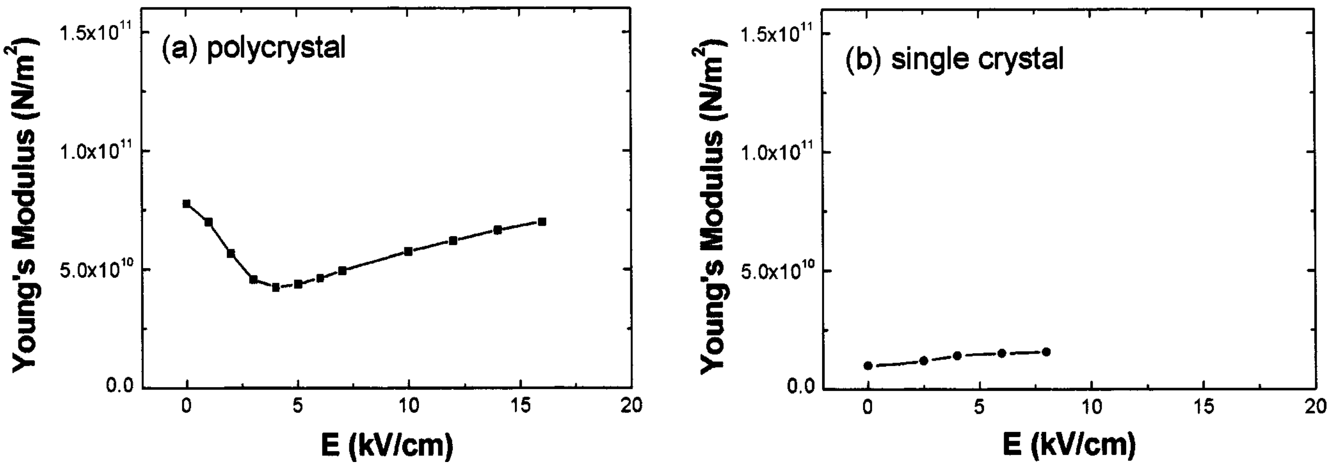

FIG. 3. Young's modulus as a function of E for single and polycrystalline PMN–PT 70/30 for $\sigma=6\times10^7$ N/m^2: (a) polycrystalline form and (b) single crystal form.

One of the major differences in the $P-E$ characteristics between the single and polycrystalline forms was the area of the hysteresis loops. Figures 2(a) and 2(b) show the hysteretic losses as a function of σ at $E=15$ kV/cm for poly- and single crystal forms, respectively. The hysteretic loss was significantly higher for the polycrystalline form, by nearly 1 order of magnitude. This dramatic difference between poly- and single crystals may be due to the multitude of randomly axed grains of the ceramic form, which results in high fractions of off-axis polarization vectors that are randomly axed with respect to E.

Young's modulus as a function of E for $\sigma=6\times10^7$ N/m^2 is shown in Figs. 3(a) and 3(b) for polycrystalline and ⟨001⟩-oriented single crystals, respectively. A dramatic difference in Y can be seen between the specimens. The value of Y was nearly 1 order of magnitude lower for the ⟨001⟩-oriented crystal, relative to the polycrystal. In addition, the dependence on E was much different for the two forms. For the ⟨001⟩-oriented single crystal, Y increased from $\sim10^{10}$ to $\sim1.5\times10^{10}$ N/m^2 with increasing E from 0 to 8 kV/cm. However, for the polycrystalline form, Y first decreased noticeably with increasing E (and also σ) and then recovered to its original value. Young's modulus was only soft in the vicinity of the coercive field (~3 kV/cm), as identified by bipolar $P-E$ measurements.

The stored elastic energy $1/2Y\epsilon^2$ was nearly the same for both the ⟨001⟩-oriented single (1.5×10^4 J/m^3) crystal and the polycrystalline (1.2×10^4 J/m^3) form. The dramatically higher value of Y indicates that the induced strains in the polycrystalline condition are noticeably lower than that of single crystals. Recent investigations of soft and hard PZTs[14] have shown similar depolarization, increase of hysteretic loss with increasing σ, and relatively high values of Y, as reported here for the PMN–PT polycrystalline form. Randomly axed grains seemingly increase hysteretic losses due to a high fraction of off-axes polarization vectors.

In ⟨001⟩-oriented PMN–PT 70/30 crystals, the stable phase under zero field is either the ferroelectric rhombohedral FE_r[1–3] or monoclinic FE_m.[15] Upon poling a FE_r or FE_m phase along the ⟨001⟩, the specimen must form a polydomain structure. Thus, both the single and polycrystalline forms of PMN–PT 70/30 have multiple domains in the poled condition. The main difference is that in single crystals all domain walls are coherent throughout the specimen and are restricted to crystallographically equivalent directions. However, in the polycrystalline form, the grains are randomly axed with respect to each other, and thus domains do not remain coherent across grain boundaries. Accordingly, only in the polycrystalline condition do hysteretic losses become pronounced.

We also measured the anisotropy of Young's modulus using the conventional resonance–antiresonance method. Temperature dependent data are shown in Fig. 4 for ⟨001⟩, ⟨110⟩, and ⟨111⟩ oriented PMN–PT 70/30 crystals. These measurements were performed under zero uniaxial stress (i.e., $\sigma=0$ N/m^2). The room temperature value of Y can be seen to be low ($\sim2\times10^{10}$ N/m^2) along both the ⟨001⟩ and ⟨110⟩ directions; whereas along the ⟨111⟩, it can be seen to be much higher ($\sim1.5\times10^{11}$ N/m^2). Previous investigations of $Pb(Zn_{1/3}Nb_{2/3})O_3-4.5\%PbTiO_3$ crystals have yielded similar anisotropic values for $Y_{\langle001\rangle}$ and $Y_{\langle111\rangle}$.[16] However, in our study, in the (011) plane, an unusual isotropy of Y was also found, where $Y_{\langle001\rangle}=Y_{\langle110\rangle}\ll Y_{\langle111\rangle}$. Averaging of anisotropic elastic constants in ceramics can lead to a large difference between Y of the ceramic and crystal forms. However, this difference will be noticeably lowered due to the

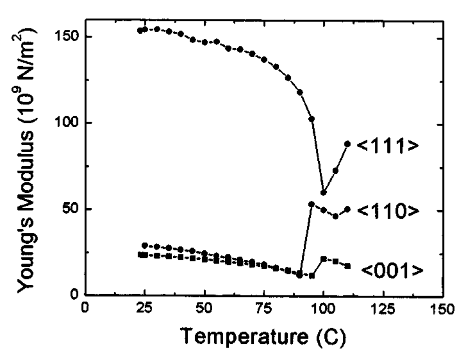

FIG. 4. Young's modulus as a function of temperature for ⟨001⟩, ⟨110⟩, and ⟨111⟩-oriented PMN–PT 70/30 single crystals for $\sigma=0$ N/m^2. These measurements were performed using the conventional IEEE resonance–antiresonance method.

unusual isotropy in the (011) plane. The large difference in Y between ceramic and crystal forms may reflect clamping of the polarization vectors of neighboring grains in the ceramic form, decreasing the ease of polarization rotation in either the FE_r or FE_m phases, and subsequently increasing the hysteretic losses.

SUMMARY

In summary, Young's modulus and hysteretic loss are much higher in polycrystalline form, relative to ⟨001⟩ or ⟨110⟩-oriented single crystal ones. For ceramics, field dependent measurements revealed a significant softening of Y, which was not observed for oriented crystals. For both forms, the elastic energy densities were similar, however significantly higher hysteretic losses were found for polycrystals.

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