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Influence of Domain Size on the Scaling Effects in Pb(Mg_{1/3}Nb_{2/3})O₃-PbTiO₃ Ferroelectric Crystals

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Abstract

The property degradation observed in thin Pb(Mg_{1/3}Nb_{2/3})O₃-PbTiO₃ (PMN-PT) crystals is believed to relate to large domains and subsequent clamping induced by surface-boundary. In this work, the properties were investigated as function of domain size, using controlled poling. The degraded piezoelectric and dielectric properties of thin PMN-PT were found to increase significantly, by decreasing domain size. Furthermore, the fine domain structure was found to be stable at 3kV/cm after 7.0×10⁵ negative-pulse cycles, hence, enabling PMN-PT crystals for high-frequency (>20 MHz) ultrasound-transducers.

Keywords

Crystalline oxides; electrical properties; dielectrics; piezoelectricity; ferroelectricity

I. Introduction

High frequency ultrasonic transducers (20–100MHz) are widely used in the area of medical diagnosis, such as dermatologic skin imaging, ophthalmic eye imaging, and catheter-based intravascular (IVUS) imaging owing to the higher imaging resolution^{1–3}. One of the main limitations for high frequency imaging, however, is a shallow depth of field caused by increased attenuation with frequency. Improvements in signal-to-noise ratio (SNR) and transducer bandwidth help overcome limitations, which can be achieved with improvements in the piezoelectric transducer^{4–7}.

Ferroelectric relaxor-PT single crystals, specifically Pb(Mg_{1/3}Nb_{2/3})O₃-PbTiO₃ (PMN-PT), offer high dielectric ($\epsilon_r > 6000$) and electromechanical coupling ($k_{33} > 0.9$), which can be used to fabricate high-sensitivity, broad-bandwidth ultrasonic transducers. However, the dielectric and piezoelectric properties of PMN-PT single crystals have been reported to significant decrease with decreasing sample thickness (increasing operational frequency), limiting its performance in high frequency ultrasound^{10,11}. It was speculated that the

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observed degradation was related to the relatively coarse domain size ($> 20\mu\text{m}$), which leads to polarization clamping as the sample thickness approaches its domain size. Therefore, it was hypothesized that finer domain PMN-PT crystals would result in improved piezoelectric and dielectric properties in the high frequency range. It has been reported that the evolution of the domain and growth of domain in ferroelectric materials can be controlled during poling with an appropriate electric field and temperature^{12,13}; thus, it can be anticipated that relatively small domain size of PMN-PT crystals can be engineered with specific poling conditions.

In this work, fine domain PMN-PT single crystals were engineered with controlled poling, in order to minimize scaling effects observed in PMN-PT crystals. In addition, the stability of the fine domain structures and electrical properties of domain-size engineered crystals were investigated as a function of pulse-field and/or with increasing number of pulses (pulse cycle).

II. Experiment

The PMN-PT crystals used in this work were grown along the [100] direction using the modified Bridgman method¹⁴. Crystal samples with an area of $5\times 5\text{ mm}^2$ were polished to a thickness of ~ 100 microns. Semi-transparent gold electrodes were sputtered on the large surfaces to allow in-situ microscopic observation of the domain structures, and to allow electrical property measurements. Poling was done by applying an electric field along the [100] direction with different temperatures. The domain structure was subsequently investigated using a polarizing light microscope (Zeiss Axioskop) with a digital camera (Pixelink) connected to computer. The dielectric permittivity was calculated from the capacitance using an HP4284A multi-frequency LCR meter at 1kHz. Unipolar strain (S-E) was measured using a modified Sawyer-Tower circuit and linear variable differential transducer (LVDT), driven by a lock-in amplifier (Stanford Research Systems, Model SR830). The high field piezoelectric coefficient (d_{33}) was determined from the slope of unipolar strain curve. An Olympus panametrics 200 MHz computer controlled pulser/receiver with a duration of 5 ns was employed to investigate domain stability.

III. Results and Discussion

Fig. 1 shows the domain structure observed in [100] oriented PMN-PT crystals developed under different poling conditions. It can be seen from Fig. 1(a) that when the crystals were poled at room temperature, laminar domains with [001] boundaries were formed. The width of the domains varied from 20 micron to 30 microns. Utilizing a field-cooling method, in which the samples were heated to 260 C with an applied electric-field of 6kV/cm, followed by cooling to room temperature at 2 C/min, the domain structure in PMN-PT crystals was found to be significantly smaller, as shown in Fig. 2(b), with size varying from the maximum of 10 micron to less than 5 micron.

The dielectric and piezoelectric properties of the sample with different domain size are reported in Table I. Also, reported are the values of thick samples with no degradation. It was found that the dielectric permittivity of fine domain PMN-PT crystals maintained a similar level (~ 6000) to the relatively thick ($\sim 1\text{ mm}$) PMN-PT crystals. In contrast, the dielectric permittivity of the coarse domain (>20 micron) PMN-PT crystals showed significantly reduced properties, being on the order of 3000, when the thickness was reduced to 100 microns.

The unipolar strain behavior for fine and coarse domain PMN-PT crystals ($\sim 100\mu\text{m}$) is presented in Fig. 2. It can be seen that the strain of the fine domain PMN-PT crystals was significantly higher than that of coarse domain crystals. The calculated high field d_{33} s from

the strain-field curves of the fine domain PMN-PT crystals were found to be about 2200 pm/V, with similar strain levels as observed in thick non-degraded PMN-PT crystals. In contrast, coarse domain PMN-PT crystals exhibited reduced strains, with piezoelectric coefficient being less than 1300 pm/V, as summarized in Table I.

The stability of the engineered domain structure was further investigated by observing the domain structure as a function of pulse-field and number of cycles on the thin PMN-PT crystals. As shown in Fig. 3(a), the domain structure of PMN-PT crystals exhibited no noticeable change in the size and shape, after $\sim 10^5$ pulse cycles with amplitude on the order of 3kV/cm, higher than E_C (~ 2 kV/cm). The domain width was found to increase slightly, being on the order of 5–20 μ m, with further increasing pulse cycles ($>10^7$), as shown in Fig. 3(b). Fig. 3(c–d) shows the change in domain size and structure with increasing pulse cycles at 6 kV/cm ($\sim 3E_C$), from which, it can be observed that the domains broadened substantially with increasing pulse cycles, being on the order of 10–40 microns, similar to those observed in room temperature poled single crystals. It is believed that the enlarged domain structure may be due to the nucleation and growth of domains with increasing number of high-energy pulses.

Fig. 4 shows unipolar strain behavior of PMN-PT crystals (thickness $\sim 100\mu$ m) at a field of 10 kV/cm, after different pulse tests, where (a–d) correspond to average domain widths of 5, 10, 20 and 30 microns, respectively. The piezoelectric coefficient d_{33} calculated from the strain-field curves was found to be ~ 2000 pm/V (Fig. 4(a)) when relatively smaller domains were present, similar to non-degraded crystals. When the domains became broader after high-energy pulses, a decrease in the maximum strain level of PMN-PT crystal was observed as demonstrated in Figures 4(b–d). A similar trend was also observed for the dielectric properties: a decrease in dielectric permittivity with corresponding increased domain size, confirming that property degradation observed in thin plates are due to the domain size effects. The piezoelectric and dielectric properties of domain-size engineered PMN-PT crystals are summarized in Table II.

IV. Summary

The dielectric and piezoelectric properties of PMN-PT crystals were found to degrade with decreasing sample thickness, owing to the large domain size ($> 20\mu$ m) associated with the scaling effects from the sample surface boundary, greatly limited their applications in high frequency range (>20 MHz). Field-cool poling approach was utilized to achieve significantly smaller domain size ($\sim 5\mu$ m), consequently leading to the improved dielectric and piezoelectric properties in thin crystal samples with thickness being on the order of <100 μ m. The degraded piezoelectric d_{33} coefficients and dielectric properties in thin samples were found to increase from 1300 pm/V to 2200pm/V and 3000 to 5600, respectively, by drastically decreasing domain size. The engineered fine domain structure of PMN-PT crystals was found to be stable up to 3kV/cm ($>E_C$) of a reverse pulse-field, after 7.0×10^5 cycles, making PMN-PT crystals promising candidates for high frequency (>20 MHz) transducer applications.

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References

1. Lockwood G, Turnbull D, Christopher D, Foster F. IEEE Eng Med Biol Mag. 1996; 15:60.
2. Turnbull D, Ramsay J, Shivji G, Bloomfield T, From L, Sauder D, Foster F. Ultrason Med Biol. 1996; 22:845.
3. Foster F, Pavlin C, Harasiewicz K, Christopher D, Turnbull D. Ultrason Med Biol. 2000; 26:1.
4. Wang H, Jiang B, Shrout T, Cao W. IEEE Trans Ultrason Ferro Freq Control. 2004; 51:908.
5. Jiang X, Snook K, Hackenberger WX. Geng, Proceeding SPIE. 2007; 6531:65310F.
6. Jiang X, Snook K, Hackenberger W, Cheng A. J Xu IEEE Sensors. 2008:573.
7. Jiang X, Snook K, Walker T, Portune A, Haber R. Proceeding SPIE. 2008; 6934:69340D.
8. Kuwata J, Uchino K, Nomura S. Jpn J Appl Phys. 1982; 21:1298.
9. Park S, Shrout T. J Appl Phys. 1997; 82:1804.
10. Lee H, Zhang S, Luo J, Li F, Shrout T. Adv Funct Mater. 2010; 20:3154.
11. Lee H, Zhang S, Shrout T. J Appl Phys. 2010; 107:124107. [PubMed: 20644658]
12. Wada S, Yako K, Kakemoto H, Tsurumi T, Kiguchi T. J Appl Phys. 2005; 98:014109.
13. Rao W, Xiao K, Cheng T, Zhou J, Wang Y. Appl Phys Lett. 2010; 97:162901.
14. www.trstechnologies.com

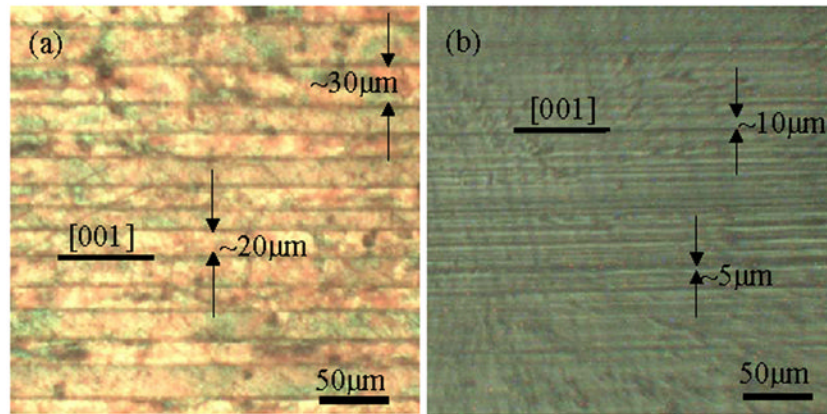


Fig. 1. Domain structure in (a) room temperature poled and (b) field-cooled [100]-oriented PMN-PT crystals.

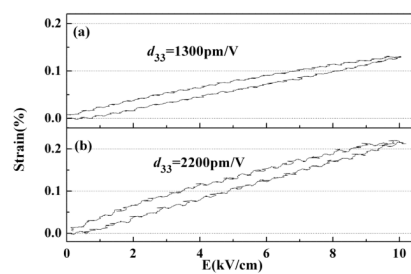


Fig. 2. Unipolar strain as a function of electric field for (a) room temperature poled and (b) field cooling [100]-oriented PMN-PT crystals (sample thickness $\sim 100\mu\text{m}$).

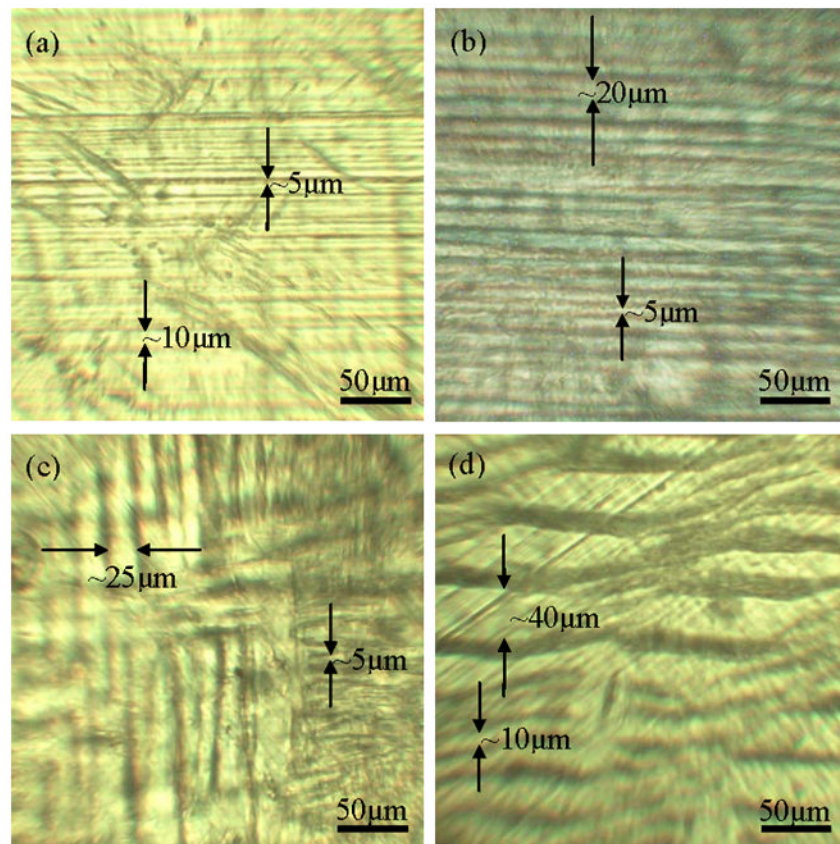


Fig. 3. Domain structure of PMN-PT crystals observed under various pulse conditions: (a) at 3kV/cm after 7.0×10^5 pulses, (b) at 3kV/cm after 7.0×10^7 pulses, (c) at 6kV/cm after 7.0×10^5 pulses, (d) at 6kV/cm after 1.5×10^6 pulses.

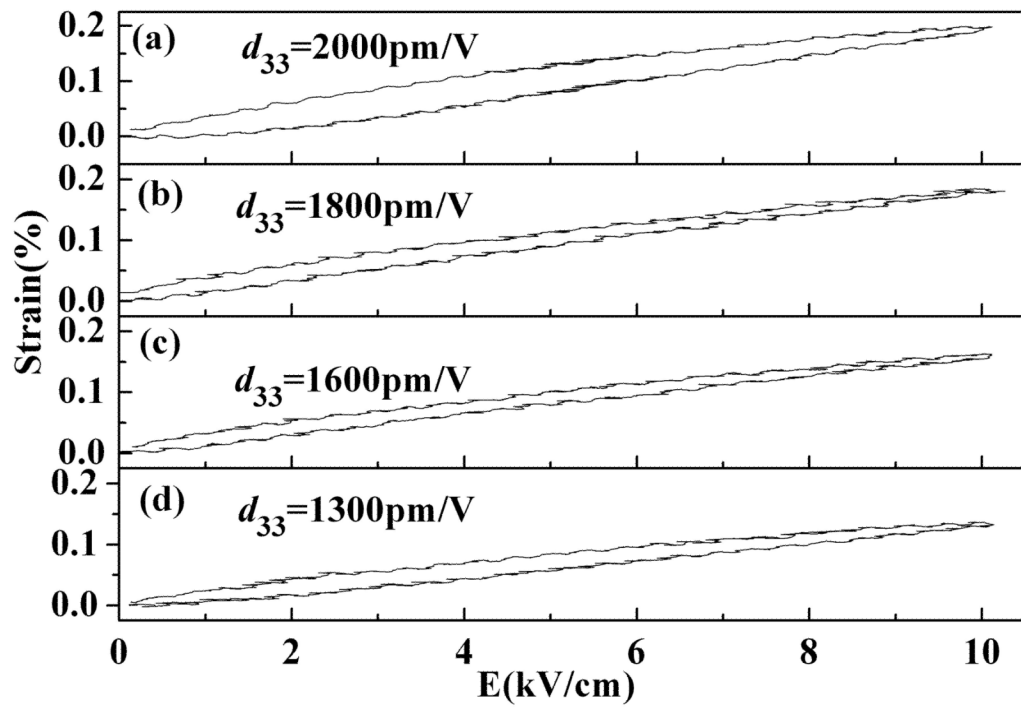


Fig. 4. Unipolar strain of PMN-PT crystals as a function of electric field after pulse tests: (a) at 3kV/cm after 7.0×10^5 pulses, (b) at 3kV/cm after 7.0×10^7 pulses, (c) at 6kV/cm after 7.0×10^5 pulses, (d) at 6kV/cm after 1.5×10^6 pulses.

Table I

The piezoelectric and dielectric properties in [100]-poled PMN-PT crystals as a function of domain size

Poling condition	Domain size (μm)	Thickness (μm)	d_{33} (pC/N)	ϵ_r
Poled at 25°C/8kV/cm	>20	1000	1800	6000
Poled at 25°C/8kV/cm	>20	100	1300	3000
Field-cooling @ 6kV/cm	~5	100	2200	5600

Piezoelectric and dielectric properties in [100]-poled domain-size engineered PMN-PT crystals, as a function of pulse field and number of pulse cycles

Table II

Pulse-field (kV/cm)	Pulse duration (ns)	Pulse cycle	Domain size (μm)	d_{33} (pm/V)	ϵ_r
Room Temperature Poling					
3	5	7.0×10^7	20~30	1300	3000
6	5	1.5×10^6	~25	1300	3000
			20~32	1300	3000
Field-cool Poling					
3	5	1.0×10^5	5~10	2200	5600
3	5	3.5×10^5	5~10	2200	5600
3	5	7.0×10^5	5~10	2000	5500
3	5	7.0×10^7	5~20	1800	5300
6	5	3.5×10^5	5~10	2000	5500
6	5	7.0×10^5	5~25	1600	4800
6	5	1.5×10^6	10~40	1300	4100