OPTOELECTRONICS AND ADVANCED MATERIALS – RAPID COMMUNICATIONS Vol. 15, No. 9-10, September-October 2021, p. 509 - 512

DC bias field effect on clamped dielectric property of $[001]_c$ poled xPb($In_{1/2}Nb_{1/2}$)O_3-(1-x-y) Pb($Mg_{1/3}Nb_{2/3}$)O_3-yPbTiO_3 single crystals

YIZHENG TANG*, QINQIN ZHONG, HANBING GU, KAI ZHANG
Hangzhou Applied Acoustics Research Institute, Hangzhou 310012, China

The relaxor-based xPb($In_{1/2}Nb_{1/2}$)O_3-(1-x-y)Pb($Mg_{1/3}Nb_{2/3}$)O_3-yPbTiO_3 (PIN-PMN-PT) single crystals have attracted a lot of attention over the last two decades due to their ultrahigh piezoelectric and electromechanical coupling properties. The elastic, dielectric, and piezoelectric properties of relaxor-based single crystals should be determined before they are used to design piezoelectric devices. Researchers can carry out simulations to evaluate the properties of piezoelectric devices after these electromechanical properties are known. To avoid the depolarization, sometimes piezoelectric devices are driven under DC bias field. Therefore, studying the dielectric property of PIN-PMN-PT single crystals under DC bias field is necessary to the design of piezoelectric devices based on them. In this study, the clamped dielectric parameters ε_{11}^S and ε_{33}^S of $[001]_c$ poled PIN-PMN-PT single crystals under DC bias field is experimentally analyzed. Moreover, it is compared with those of lead zirconate titanate (PZT).

(Received February 28, 2021; accepted October 7, 2021)

Keywords: Relaxor-based single crystals, PIN-PMN-PT, DC bias, Clamped dielectric property

1. Introduction

Relaxor-based single crystals have great potential for use in both sensor and actuator applications [1–4] because of their excellent piezoelectric properties and high electromechanical coupling coefficients at compositions near the morphotropic phase boundary (MPB). The first generation relaxor-based single crystals, such as PZN-PT and PMN-PT, have a low Curie temperature and a low rhombohedral–tetragonal phase-transition temperature, which limit their application in transducers [4]. Compared with the binary PMN-PT and PZN-PT single crystals, the second generation relaxor-based single crystals, such as the ternary Pb($In_{1/2}Nb_{1/2}$)O_3-Pb($Mg_{1/3}Nb_{2/3}$)O_3-PbTiO_3 (PIN-PMN-PT) single crystals, have higher phase transition temperatures and larger coercive field near MPB compositions [5–8]. Therefore, they extend the high electromechanical properties to a broader range of temperature, electric field and mechanical stress. Note that the superior longitudinal piezoelectric constants and excellent electromechanical coupling factors appear only in the $[001]_c$ poled rhombohedral PMN–PT, PZN–PT and PIN-PMN-PT single crystals [2].

The elastic, dielectric, and piezoelectric properties of relaxor-based single crystals should be determined before they are used to design piezoelectric devices [9–11]. Researchers can carry out simulations to evaluate the properties of piezoelectric devices after these electromechanical properties are known. In many practical applications, piezoelectric devices are driven under strong electric field or under DC bias field [12]. For example, piezoelectric materials used in high power transmitting transducers may depolarize after a long time work. Strong DC bias field can help piezoelectric materials avoid depolarization. Therefore, studying the dielectric property of PIN-PMN-PT single crystals under DC bias field is necessary to the design of piezoelectric devices based on them. Wang et al. [12] studied the effect of DC bias field on the complex materials coefficients of a thin plate soft PZT resonator. Masys et al. [13] investigated the electromechanical response of piezoelectric ceramics as a function of the amplitude and frequency of large electric fields and studied the effects of dc bias fields. Zhang et al. [14] studied the dielectric properties and DC bias characteristic of a relaxor ferroelectric ceramic system of Pb($Mg_{1/2}W_{1/2}$)O_3-PbTiO_3-Pb($Ni_{1/3}Nb_{2/3}$)O_3. Shen et al. [15] studied the DC field effect on dielectric property of Ba(Zr_xTi_{1-x})O_3 ceramics. Wan et al. [16] studied the temperature and DC bias dependence of the phase transition behavior of [011]- and [001]-oriented PIN–PMN–PT single crystals with MPB composition below 100 kHz.

In this study, the clamped dielectric parameters ε_{11}^{S} and ε_{33}^{S} of $[001]_c$ poled 30PIN-40PMN-30PT single crystals under DC bias field are measured. Moreover, they are compared with those of lead zirconate titanate (PZT). The influence of DC bias field on the clamped dielectric property of $[001]_c$ poled 30PIN-40PMN-30PT single crystals is analyzed.

2. Materials and methods

The experimental 30PIN-40PMN-30PT single crystal was grown by the Bridgman technique. The as-grown crystal sample has the growth direction along $[111]_c$. The sample orientation $[001]_c$ was determined by the X-ray single crystal orientation devices. The sample was poled along $[001]_c$ under 1kV/mm DC field and 140 °C temperature after the electrodes on the $(001)_c$ surfaces were sputtered. Two rectangular parallelepiped $[001]_c$ poled 30PIN-40PMN-30PT single crystal samples were prepared from different batches of products. The geometrical parameters of sample #1 are 2.300 × 3.663 × 4.021 mm^3. Its density determined according to the volume and mass is 8104 kg/m^3. The geometrical parameters of sample #2 are 2.836 × 4.237 × 4.023 mm^3. Its density is 8184 kg/m^3. For comparison, a rectangular parallelepiped PZT-8 sample is prepared. The PZT-8 sample was purchased from Yu Hai Electric Ceramics Co., Ltd. (China). Its sizes and density are 5.593 × 4.710 × 3.371 mm^3 and 7670 kg/m^3, respectively.

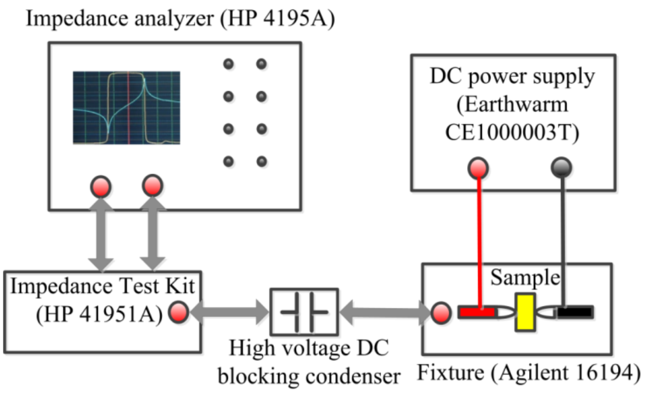

Fig. 1. Measurement setup for dielectric parameters under DC bias (color online)

Fig. 1 shows the measurement setup for clamped dielectric parameters under DC bias field, which is composed of an impedance analyzer (HP 4195A), an impedance test kit (HP 41951A), a fixture (Agilent 16194), a high voltage DC blocking condenser and a DC power supply (Earthwarm CE10000eT). The high voltage DC blocking condenser was used to protect the impedance analyzer. The high frequency capacitance C_h of the sample was measured at 35 MHz. Before measurement, the impedance test kit should be calibrated and the fixture should be compensated. Note that the fixture compensation should be carried out after the DC power supply is connected to the fixture, otherwise, the DC power supply may bring measurement error. Moreover, the direction of the electric field intensity is along the polarization direction during the measurement of capacitance along this direction. The relative clamped dielectric parameter ε_{33}^{S} of the sample can be obtained using $\varepsilon_{33}^{S} = C_h t/(A\varepsilon_0)$, where t is the sample thickness, A is the area of the sample surface perpendicular to the polarization direction, and ε_0 is the permittivity of vacuum.

3. Results and discussion

Fig. 2 shows the relative clamped dielectric parameters ε_{11}^{S} and ε_{33}^{S} under DC bias field of two $[001]_c$ poled 30PIN-40PMN-30PT samples. The solid red and dashed blue lines are corresponding to samples #1 and #2, respectively. The relative variation of ε_{11}^{S} between samples #1 and #2 is about 12%, while that of ε_{33}^{S} between samples #1 and #2 is about 1.6% when there is no DC bias field. This indicates that the property difference between different batches of $[001]_c$ poled 30PIN-40PMN-30PT single crystals is significant. So far, the homogeneity problem of different batches of relaxor based single crystals has not been perfectly solved. The quality of single crystals is closely related to the growth technique. The following techniques are often used to grow relaxor-based ferroelectric single crystals: the conventional flux technique [17], the Bridgman technique [18], the modified Bridgman techniques [19], the top-seeded solution growth (TSSG) technique [20], and the solid-state crystal growth technique [21]. However, growing relaxor-based ferroelectric single crystals with a large size, high piezoelectric quality, and high homogeneity is one of the major challenges that must be overcome.

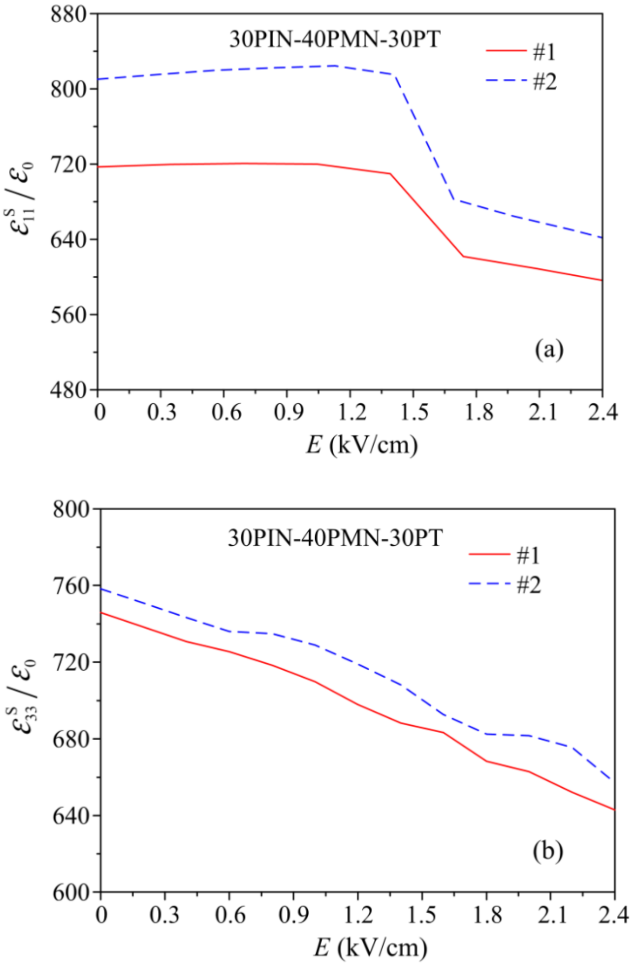

Fig. 2. Clamped dielectric parameters under DC bias field of 30PIN-40PMN-30PT samples. (a) $\varepsilon^S_{11}/\varepsilon_0$*; (b)* $\varepsilon^S_{33}/\varepsilon_0$ *(color online)*

Fig. 2(a) shows that the clamped dielectric parameter ε^S_{11} of the $[001]_c$ poled 30PIN-40PMN-30PT single crystal is insensitive to the variation of DC bias field E when it is less than 1.3 kV/ cm. However, the value of ε^S_{11} drops sharply when E reaches about 1.3 kV/cm, then it decreases slowly when E is greater than 1.7 kV/cm. The sharp drop between 1.3 kV/cm and 1.7 kV/cm probably be triggered by the variation of electric domain which is caused by the external electric field. Fig. 2(b) indicates that DC bias field has a great influence on the clamped dielectric parameter ε^S_{33} of the $[001]_c$ poled 30PIN-40PMN-30PT single crystal. ε^S_{33} decreases with an increase in E. It decreases about 6.4% and 13.8% when E reaches 1.2 kV/cm and 2.4 kV/cm, respectively. The variation of the clamped dielectric parameters may lead to the shift of the electric resonance and anti-resonance frequencies. Therefore, to improve the transmitting efficiency of the transducers made by PIN-PMN-PT single crystals, the aforementioned influence should be considered in the design of the power-driven circuit of the transducers when they work under a high DC bias field.

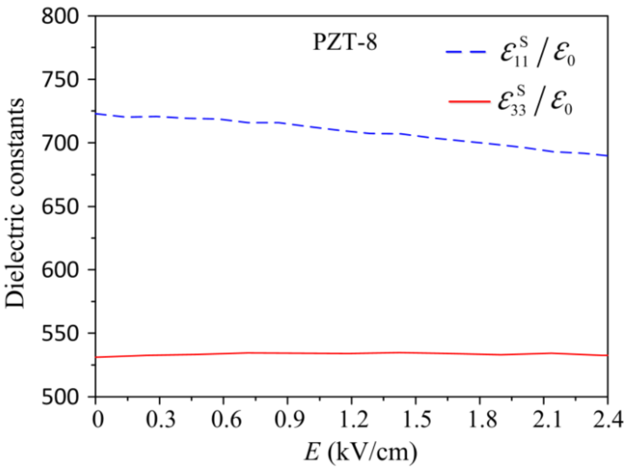

Fig. 3. Clamped dielectric parameters under DC bias field of PZT-8 sample (color online)

Fig. 3 shows the relative clamped dielectric parameters ε^S_{11} and ε^S_{33} under DC bias field of the PZT-8 sample. ε^S_{33} is very stable under DC bias field. The relative variation between the ε^S_{33} under E = 0 kV/cm and that under E = 1.2 kV/cm is only about 0.6%. ε^S_{11} decreases slowly with an increase in E. The relative variation between the ε^S_{33} under E = 0 kV/cm and that under E = 1.2 kV/cm is about 2.1%. Compared with $[001]_c$ poled 30PIN-40PMN-30PT single crystals, PZT-8 has more stable clamped dielectric parameters under strong DC bias field.

4. Conclusions

In this study, the clamped dielectric property of $[001]_c$ poled PIN-PMN-PT single crystals under DC bias field is studied. Experimental results show that DC bias field has a great influence on their clamped dielectric property. For example, ε^S_{33} decreases about 6.4% and 13.8% when E reaches 1.2 kV/cm and 2.4 kV/cm, respectively. Moreover, a sharp drop of ε^S_{11} occurs between 1.3 kV/cm and 1.7 kV/cm, which probably be caused by the variation of electric domain.

Compared with traditional PZT, $[001]_c$ poled PIN-PMN-PT single crystals have far higher piezoelectric and electromechanical coupling properties. However, their material properties are more complex and unstable, for example, their dielectric parameters are sensitive to the variation of DC bias field. Therefore, the designers of piezoelectric transducers should carefully choose piezoelectric materials according to requirements. To improve the transmitting efficiency of the transducers made by PIN-PMN-PT single crystals, the power-driven circuit should consider the influence of DC bias field.

Acknowledgments

The authors expresse their sincere gratitude to Mr. M. Dong and Mr. L. Zhong for their support during the experiment. The author also thanks to M. Wu for his help in the preparation of the manuscript.

References

[1] S. E. Park, T. R. Shrout, J. Appl. Phys. **82**, 1804 (1997).

[2] E. W. Sun, W. W. Cao, Prog. Mater. Sci. **65**, 124 (2014).

[3] S. J. Zhang, F. Li, J. Appl. Phys. **111**, 031301 (2012).

[4] S. J. Zhang, F. Li, X. N. Jiang, J. Kim, J. Luo, X. C. Geng, Prog. Mater. Sci. **68**, 1 (2015).

[5] H. C. Song, J. Y. Ha, J. S. Kim, S. J. Yoon, D. Y. Jeong, Jpn. J. Appl. Phys. **46**, 1540 (2007).

[6] F. M. Wu, B. Yang, E. W. Sun, G. Liu, H. Tian, W. W. Cao, J. Appl. Phys. **114**, 027021 (2013).

[7] E. W. Sun, W. W. Cao, P. D. Han, Mater. Lett. **65**, 2855 (2011).

[8] X. Z. Liu, S. J. Zhang, J. Luo, T. R. Shrout, W. W. Cao, J. Appl. Phys. **106**, 074112 (2009).

[9] S. Saitoh, T. Kobayashi, K. Harada, S. Shimanuki, Y. Yamashita, IEEE Trans. Ultrason. Ferroelect. Freq. Contr. **46**, 152 (1999).

[10] W. Zhang, X. M. Li, R. Zhang, W. W. Cao, Chin. Phys. Lett. **26**, 064301 (2009).

[11] S. T. Lau, H. Li, K. S. Wong, Q. F. Zhou, D. Zhou, Y. C. Li, H. S. Luo, K. K. Shung, J. Y. Dai, J. Appl. Phys. **105**, 094908 (2009).

[12] Q. M. Wang, T. Zhang, Q. M. Chen, X. H. Du, Sensor. Actuat. A-Phys. **109**, 149 (2003).

[13] A. J. Masys, W. Ren, G. Yang, B. K. Mukherjee, J. Appl. Phys. **94**, 1155 (2003).

[14] Y. Zhang, L. T. Li, Z. L. Gui, J. M. Tian, Fermelectrics, **247**, 259 (2000).

[15] B. Shen, Q. W. Zhang, J. W. Zhai, Z. K. Xu, Ceram. Int. **39**, S9 (2013).

[16] Y. H. Wan, Z. R. Li, M. Ma, S. J. Fan, Z. Xu, J. Mater. Res. **33**, 4053 (2018).

[17] S. E. Park, M. L. Mulvihill, G. Risch, T. R. Shrout, Jpn. J. Appl. Phys. **36**, 1154 (1997).

[18] P. Yu, F. F. Wang, D. Zhou, W. W. Ge, X. Y. Zhao, H. S. Luo, J. L. Sun, X. J. Meng, J. H. Chu, Appl. Phys. Lett. **92**, 252907 (2008).

[19] X. M. Wan, J. Wang, H. L. W. Chan, C. L. Choy, H. S. Luo, Z. W. Yin, J. Cryst. Growth **263**, 251 (2008).

[20] W. Chen, Z. G. Ye, J. Cryst. Growth **233**, 503 (2001).

[21] S. J. Zhang, S. M. Lee, D. H. Kim, H. Y. Lee, T. R. Shrout, J. Am. Ceram. Soc. **91**, 683 (2008).

---

[*] Corresponding author: tyz361@163.com