2012

An efficient way to enhance output strain for shear mode Pb(In1/2Nb1/2)O-3-Pb(Mg1/3Nb2/3)O-3-PbTiO3 crystals: Applying uniaxial stress perpendicular to polar direction

Fei Li
Xi'an Jiaotong University, lifei1216@gmail.com

Shujun Zhang
Xi'an Jiaotong University, shujun@uow.edu.au

Zhuo Xu
Xi'an Jiaotong University

Dabin Lin
Xi'an University of Technology

JunJie Gao
Xi'an Jiaotong University

See next page for additional authors

Publication Details

Li, F., Zhang, S., Xu, Z., Lin, D., Gao, J., Li, Z. & Wang, L. (2012). An efficient way to enhance output strain for shear mode Pb(In1/2Nb1/2)O-3-Pb(Mg1/3Nb2/3)O-3-PbTiO3 crystals: Applying uniaxial stress perpendicular to polar direction. Applied Physics Letters, 100 (19), 192901-1-192901-4.

An efficient way to enhance output strain for shear mode Pb(In1/2Nb1/2)O-3-Pb(Mg1/3Nb2/3)O-3-PbTiO3 crystals: Applying uniaxial stress perpendicular to polar direction

Abstract

The shear piezoelectric behavior of [001] poled tetragonal and [011] poled rhombohedral Pb(In1/2Nb1/2)O3-Pb(Mg1/3Nb2/3)O3-PbTiO3 (PIN-PMN-PT) crystals, with "1T" and "2R" domain configurations, respectively, were investigated under uniaxial stress perpendicular to polar direction. The shear piezoelectric coefficient d15 was found to decrease with increasing compressive stress for both "1T" and "2R" crystals. Based on thermodynamic analysis, the phase structure can be stabilized by applying compressive stress perpendicular to polar direction, resulting in a "harder" polarization rotation process, accounts for the reduced shear piezoelectric coefficient. Of particular importance is that the allowable drive electric field was greatly increased and transverse dielectric loss was drastically reduced under compressive stress, leading to the improved maximum-shear-strain.

Disciplines

Engineering | Physical Sciences and Mathematics

Publication Details

Li, F., Zhang, S., Xu, Z., Lin, D., Gao, J., Li, Z. & Wang, L. (2012). An efficient way to enhance output strain for shear mode Pb(In1/2Nb1/2)O-3-Pb(Mg1/3Nb2/3)O-3-PbTiO3 crystals: Applying uniaxial stress perpendicular to polar direction. Applied Physics Letters, 100 (19), 192901-1-192901-4.

Authors

Fei Li, Shujun Zhang, Zhuo Xu, Dabin Lin, JunJie Gao, Zhenrong Li, and Linghang Wang

An efficient way to enhance output strain for shear mode $Pb(In_{1/2}Nb_{1/2})O_3$-$Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ crystals: Applying uniaxial stress perpendicular to polar direction

Fei Li, Shujun Zhang, Zhuo Xu, Dabin Lin, JunJie Gao, Zhenrong Li, and Linghang Wang

Citation: Appl. Phys. Lett. **100**, 192901 (2012); doi: 10.1063/1.4712129
View online: https://doi.org/10.1063/1.4712129
View Table of Contents: http://aip.scitation.org/toc/apl/100/19
Published by the American Institute of Physics

An efficient way to enhance output strain for shear mode $Pb(In_{1/2}Nb_{1/2})O_3$-$Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ crystals: Applying uniaxial stress perpendicular to polar direction

Fei Li,[1,a)] Shujun Zhang,[1,2,b)] Zhuo Xu,[1] Dabin Lin,[1] JunJie Gao,[1] Zhenrong Li,[1] and Linghang Wang[1]
[1]*Electronic Materials Research Laboratory, Key Laboratory of the Ministry of Education and International Center for Dielectric Research, Xi'an Jiaotong University, Xi'an 710049, China*
[2]*Materials Research Institute, Pennsylvania State University, University Park, Pennsylvania 16802, USA*

(Received 8 March 2012; accepted 21 April 2012; published online 7 May 2012)

The shear piezoelectric behavior of [001] poled tetragonal and [011] poled rhombohedral $Pb(In_{1/2}Nb_{1/2})O_3$-$Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PIN-PMN-PT) crystals, with "1T" and "2R" domain configurations, respectively, were investigated under uniaxial stress perpendicular to polar direction. The shear piezoelectric coefficient d_{15} was found to decrease with increasing compressive stress for both "1T" and "2R" crystals. Based on thermodynamic analysis, the phase structure can be stabilized by applying compressive stress perpendicular to polar direction, resulting in a "harder" polarization rotation process, accounts for the reduced shear piezoelectric coefficient. Of particular importance is that the allowable drive electric field was greatly increased and transverse dielectric loss was drastically reduced under compressive stress, leading to the improved maximum-shear-strain. [http://dx.doi.org/10.1063/1.4712129]

Relaxor-$PbTiO_3$ (PT) crystals were reported to possess ultrahigh piezoelectric response ($d_{33} > 1500$ pC/N and $k_{33} > 90$) for compositions around morphotropic phase boundaries (MPBs),[1] attracted considerable attentions over the last 20 years, with emphasis on the fundamental and application researches.[2–8] The relationship between piezoelectric properties and phase/domain structures has been extensively studied in relaxor-PT crystals, e.g., $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PMN-PT) and $Pb(In_{1/2}Nb_{1/2})O_3$-$Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PIN-PMN-PT), in order to explore the origin of the high longitudinal piezoelectric activity,[2–5] which was believed to be intrinsic (lattice deformation) in nature. The ultrahigh single domain shear piezoelectric activity is thought to be the dominant factor for the intrinsic contribution in relaxor-PT crystals.[4,5]

From application viewpoint, shear piezoelectric response of relaxor-PT crystals drew attentions for transducer design, due to the high piezoelectric coefficient d_{15} ($\sim$2000–7000 pC/N), electromechanical coupling $k_{15} > 90\%$ and elastic compliance s_{55}^E ($\sim$100–250 pm^2/N), allowing for design of ultralow frequency and broad bandwidth transducers with minimized dimension.[9–15] In addition, a temperature independent shear piezoelectric response, together with ultrahigh value $d_{24} > 2000$ pC/N, has been achieved for [011] poled orthorhombic crystals in the temperature range of $-50\sim100\,^\circ$C, due to the vertical orthorhombic-rhombohedral phase boundary.[16,17] For comparison, the variation of longitudinal piezoelectric coefficient is up to 300% for relaxor-PT crystals in the same temperature range, due to the curved rhombohedral-tetragonal phase boundary.

However, the main drawback of the thickness shear-mode crystals is the low allowable ac electric field. At low electric field, the shear piezoelectric response is related to a polarization rotation process, which is a reversible process, as shown in Fig. 1(a), while at high electric field, irreversible process happens with occurrence of new domains, corresponding to the domain switching process, as shown in Fig. 1(b). Due to the appearance of new domains, the shear piezoelectric response drastically decreases and some new vibration modes (such as 31-mode) can be observed.[18] From previous investigations, the allowable ac drive electric fields of thickness shear-mode samples for relaxor-PT crystals were less than half of their respective coercive fields ($\leq$2.5 kV/cm for PIN-PMN-PT crystals), limiting the output shear strain and acoustic power.[18] Thus, it is required to enhance the allowable ac electric field for thickness shear-mode relaxor-PT crystals. The developed internal bias field (0.5$\sim$1.5 kV/cm) in Mn-doped relaxor-PT crystals were reported to increase the allowable ac drive field, being on the order of $\sim$70% of their respective coercive fields, due to the fact that the internal bias field can stabilize the domains and make the domain switching harder.[18]

Similar to the internal bias, ferroelectric domains can also be stabilized by applying dc bias electric field along polar direction or uniaxial stress perpendicular to polar

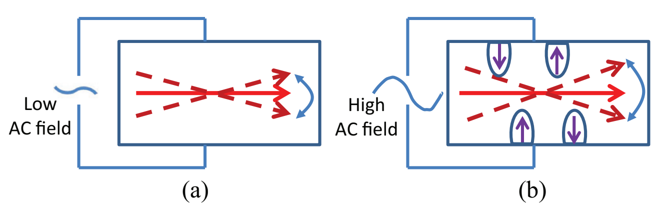

FIG. 1. Schematic polarization variation of shear-mode tetragonal crystals, where the arrows represent polar directions. (a) At low ac electric field, only reversible polarization rotation exist; (b) At high ac electric field, both polarization rotation and irreversible domain switch occur, thus new domains form (the purple arrows represent the spontaneous polarization of new domains).

a)Electronic mail: lifei1216@gmail.com.
b)Electronic mail: soz1@psu.edu.

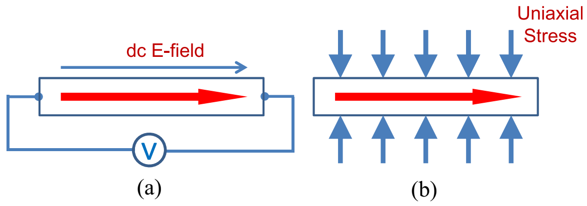

FIG. 2. Stabilization of polarization by applying (a) dc electric field and (b) compressive stress, where the red arrows represent polar directions.

direction. To stabilize ferroelectric domains, the dc electric field is required to apply along the poling direction, other than the operational direction of the shear-mode crystals, thus two pairs electrodes are needed, as shown in Fig. 2(a). However, it is convenient to apply uniaxial stress perpendicular to polar direction for shear-mode crystals, because the preload stress is along the same direction of ac drive field, as shown in Fig. 2(b). In this paper, the shear piezoelectric properties of [001] poled tetragonal and [011] poled rhombohedral PIN-PMN-PT crystals, with single domain ("1T") and multidomain ("2R") configurations, respectively, were investigated under the uniaxial compressive stress.

PIN-PMN-PT single crystals were grown by the modified Bridgman technique. The crystals were oriented by x-ray orientation system and cut to obtain shear-mode samples with dimensions of 10 mm × 10 mm × 1 mm. All the samples were electroded by vacuum sputtered gold on the polished side faces for poling process, where the [011] oriented rhombohedral crystals were poled by applying a dc field of 10 kV/cm at room temperature, while the [001] oriented tetragonal crystals were poled through Curie temperature at small electric field to avoid cracking.[10] The electrodes were removed after poling process and re-electroded on the large surfaces, being (100) and (0-11) for [001] poled tetragonal and [011] poled rhombohedral crystals, respectively.

The uniaxial stress was applied to the shear samples by a special home-made setup, with the details given in Ref. 19. In this setup, the uniaxial stress is applied to the sample via a brass holder. To minimize the friction between the sample and brass holder surfaces, the surfaces of brass holder were polished using 500 nm Al_2O_3 powder slurry to achieve mirror faces (it should be noted that there is still a possibility of some clamping associated with the fixture, even though the brass surfaces have been highly polished). The stress dependence of transverse dielectric constant $\varepsilon_{11}/\varepsilon_0$ and polarization-electric field (P-E) behaviors were measured by the LCR meter (HP4284A) and modified Sawyer-Tower circuit, respectively. At zero uniaxial stress, the shear piezoelectric coefficients were measured by impedance method following IEEE Standard, while under various uniaxial stresses, the shear piezoelectric coefficients were evaluated from the transverse dielectric permittivity using equation $d_{15} = P_3 Q_{55} \varepsilon_{11}$, where P_3 is the spontaneous polarization and Q_{55} the electrostrictive constant, ε_{11} transverse dielectric permittivity. According to thermodynamic analysis and experimental results, the variation of P_3 and Q_{55} can be neglected when compared to the dielectric permittivity, with uniaxial stress being in the range of 0∼60 MPa.[20–22] Therefore, the variation of shear piezoelectric coefficient as a function of compressive stress can be determined from the stress dependent transverse dielectric permittivity.

The general properties of the studied PIN-PMN-PT crystals were listed in Table I. The shear piezoelectric coefficients d_{15} are on the order of 2100 and 3000 pC/N for [001] poled tetragonal (1T) and [011] poled rhombohedral (2R) crystals. The high level of shear piezoelectricity of relaxor-PT crystals can be attributed to the high level of phase structural instability.[4,10] Fig. 3 showed the piezoelectric coefficient d_{15} as a functional of compressive stress perpendicular to polarization direction for "1T" and "2R" crystals. The shear piezoelectric responses were found to decrease with increasing the compressive stress, where the coefficient d_{15} decreased from 2100 pC/N to 1600 pC/N upon stress increasing from 0 to 25 MPa for "1T" crystals, while the coefficient d_{15} decreased from 3000 pC/N to 2600 pC/N upon stress increasing from 0 to 55 MPa for "2R" crystals. It should be noted that higher compressive stress could induce cracks in shear-mode PIN-PMN-PT crystals (40 MPa and 70 MPa for studied "1T" and "2R" crystals, respectively). The observed reduction of shear coefficients can be analyzed from the respect of polarization rotation.

Taking tetragonal crystal as example, the stability of phase under a compress stress can be analyzed using the free energy equation,[23]

$$\begin{aligned} G(X) = G_0 &- 1/2 s_{11}(X_1^2 + X_2^2 + X_3^2) \\ &- s_{12}(X_1X_2 + X_2X_3 + X_3X_1) \\ &- 1/2 s_{44}(X_4^2 + X_5^2 + X_6^2) \\ &- Q_{11}(X_1P_1^2 + X_2P_2^2 + X_3P_3^2) \\ &- Q_{12}[X_1(P_2^2 + P_3^2) + X_2(P_3^2 + P_1^2) + X_3(P_1^2 + P_2^2)] \\ &- Q_{44}(X_4P_2P_3 + X_5P_3P_1 + X_6P_1P_2), \qquad (1) \end{aligned}$$

where G_0 is the Gibbs free energy under zero stress, P_i the polarization, X_λ the stress, $s_{\lambda\mu}$ the elastic constants, and $Q_{\lambda\mu}$ the electrostrictive constants. For [001] poled tetragonal crystals, $P_1 = P_2 = 0$ and $P_3 \neq 0$. Under perpendicular stress shown in Fig. 3(a) ($X_1 < 0$; $X_\lambda = 0$, $\lambda = 2\sim6$), the free energy can be rewritten as follows:

$$G(X_1) = G_0 - \frac{1}{2} s_{11} X_1^2 - Q_{12} X_1 P_3^2, \qquad (2)$$

where $s_{11} > 0$ and $Q_{12} < 0$,[22,23] so $G(X_1) < G_0$, indicating that the spontaneous polarization is more stable under

TABLE I. Properties of "1T" and "2R" shear-mode PIN-PMN-PT crystals.

Material	T_c (°C)	$T_{R/O\text{-}T}$ (°C)	E_C (kV/cm)	$\varepsilon_{11}{}^T/\varepsilon_0$	$s_{55}{}^E$ (pm^2/N)	d_{15} (pC/N)	k_{15}
PIN-PMN-PT (2R)	165	125	5.0	6500	160	3000	0.92
PIN-PMN-PT (1T)	205	−10	6.0	16000	52	2100	0.85

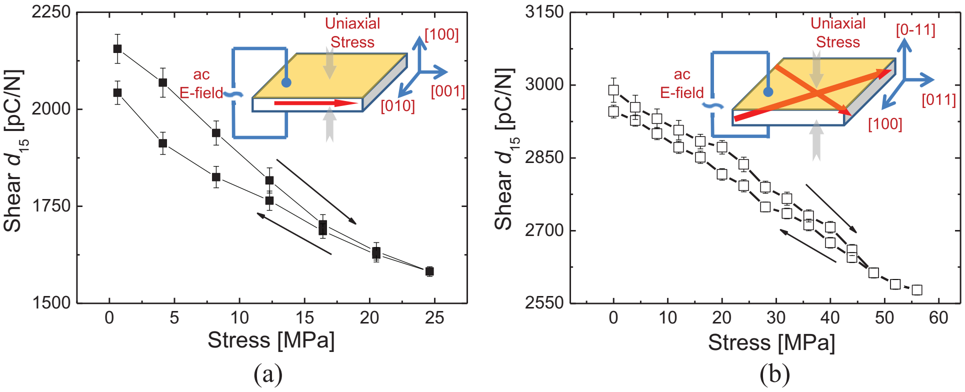

FIG. 3. Shear piezoelectric coefficients of PIN-PMN-PT crystals with respect to uniaxial compressive stress perpendicular to the polar direction (measured at 1 kHz), (a) "1T" crystals and (b) "2R" crystals. The small insets showed the schematic figures of shear samples (red arrows represent polar directions), applied stresses, and electric fields.

compressive stress when compared to zero stress condition,[20] leading to a "harder" polarization rotation process, accounts for the decrease of shear piezoelectric coefficient d_{15}.[4,10]

Figs. 4 and 5 showed the P-E curves as a function of compressive uniaxial stress for "1T" and "2R" shear-mode crystals, respectively. At low level of electric field (1 kV/cm), the polarization versus electric field followed a linear behavior for both "1T" and "2R" crystals under various uniaxial stresses, as shown in Figs. 4(a) and 5(a). This indicates that the irreversible process (i.e., domain wall motion) is minimal at low level of electric field for "1T" and "2R" crystals. In addition, it can be observed from Figs. 4(a) and 5(a) that the slope of P-E curves decreased with increasing compressive stress, again demonstrating that the transverse dielectric permittivity decreased with stress increasing.

The P-E curves of high electric field (>2.5 kV/cm) became nonlinear and hysteretic under zero uniaxial stress for both "1T" and "2R" crystals, as shown in Figs. 4(b) and 5(b), due to the occurrence of irreversible domain switching. The crystals can be partially depolarized and new domains form with further increasing the ac electric field, as illustrated in Fig. 1(b). At this condition, the nonlinear and hysteretic characteristics became more obvious, as shown in Figs. 4(c) and 5(c), thus the maximum allowable drive electric field was found to be lower than 2.5 kV/cm for both "1T" and "2R" crystals without applying the stress.

Of particular significance is that the hysteretic properties of both "1T" and "2R" crystals were greatly reduced by applying compressive stress to crystals, owing to the stabilized domains, as shown in Figs. 4 and 5. Therefore, the allowable electric fields can be enhanced by applying uniaxial compressive stress perpendicular to poling direction, which were found to increase to 4.5 kV/cm and 4 kV/cm for "1T" crystals at compressive stress of 25 MPa and "2R" crystals at 55 MPa, respectively, as listed in Table II.

With applying compressive stress, though the shear piezoelectric response decreased, the level of allowable drive field increased. Therefore, the improvement in maximum-shear-strain is expected by applying compressive stress. The maximum-shear-strain (S_{max}) is estimated by the following equation:

$$S_{max} = d_{15}(X)E_{allow}(X) \tag{3}$$

where $d_{15}(X)$ is the shear piezoelectric coefficient under a stress, $E_{allow}(X)$ the allowable electric field under a stress. As listed in Table II, the S_{max} of "1T" crystals was increased by 38% under compressive stress of 25 MPa (from 0.5‰ to 0.7‰), while the S_{max} of "2R" crystals was increased by 50% under compressive stress of 55 MPa (from 0.7‰ to 1.0‰).

The dielectric losses were calculated from the P-E loops,[8,24] as listed in Table III. It can be seen that the dielectric loss was greatly reduced by applying the compressive stress, especially at high electric field. The dielectric loss factors were found to be 20% and 50% for "2R" and "1T" crystals, respectively, at electric field of 4 kV/cm, without applying stress, being reduced to the order of 4% by applying compressive stresses of 55 MPa and 25 MPa for "2R"

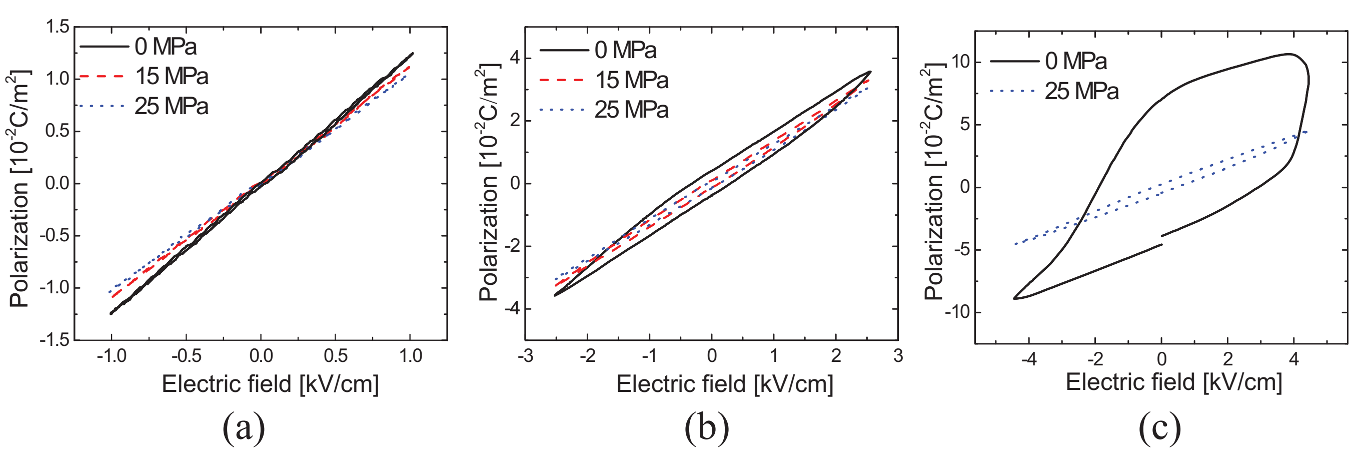

FIG. 4. P-E loops at various uniaxial stresses for "1T" PIN-PMN-PT crystals. (a) 1 kV/cm, (b) 2.5 kV/cm, and (c) 4.5 kV/cm. Measured at 1 Hz.

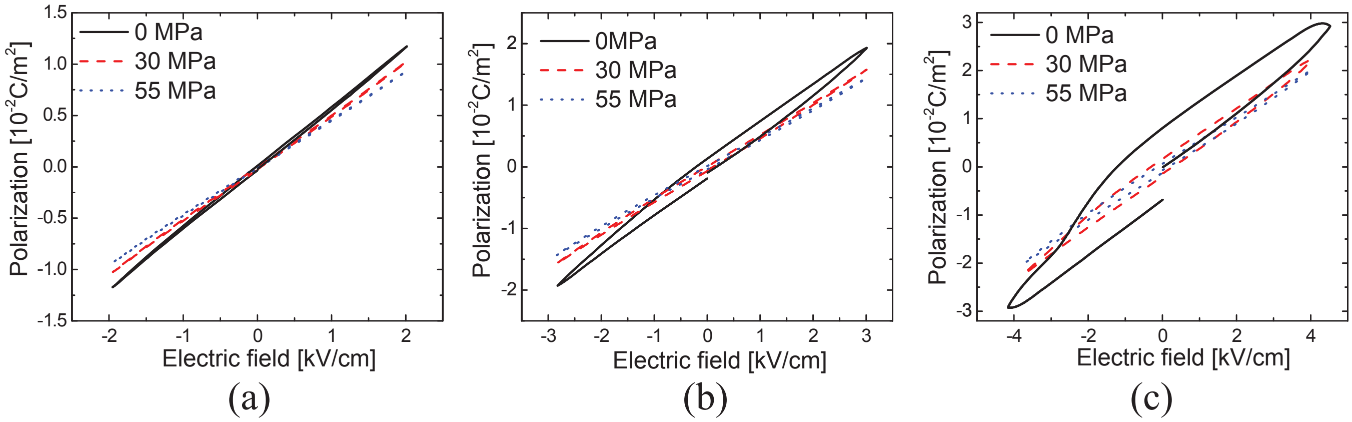

FIG. 5. P-E loops at various uniaxial stresses for "2R" PIN-PMN-PT crystals. (a) 1 kV/cm, (b) 3 kV/cm, (c) 4 kV/cm. Measured at 1 Hz.

and "1T" crystals, respectively, which will benefit the general applications, with the requirement of the loss factors being lower than 5%. Thus, the results again indicated that the allowable drive electric field of the shear-mode crystals can be enhanced by applying compressive stress perpendicular to the polar direction.

In summary, the shear piezoelectric behaviors of [001] poled tetragonal and [011] poled rhombohedral PIN-PMN-PT crystals were investigated under compressive stress perpendicular to polar direction. The shear piezoelectric coefficient d_{15} and dielectric loss were found to decrease with increasing the compressive stress, due to a "harder" polarization rotation process under compressive stress. Of particular interest is that the allowable drive electric field and maximum-shear-strain of PIN-PMN-PT crystals can be enhanced by applying compressive stress. These results will benefit the design of shear-mode transducers where high power is required.

TABLE II. Allowable drive electric field, shear piezoelectric coefficient, and maximum strain of PIN-PMN-PT crystals as a function of compressive stress. It should be noted that the shear strain cannot be estimated by Eq. (3) when the level of loss factor (or hysteresis) is relatively high. The high level of loss/hysteresis is induced by the nucleation of new domains or motion of domain walls at large ac electric field.

	"1T" crystal			"2R" crystal		
Compressive stress	0 MPa	15 MPa	25 MPa	0 MPa	30 MPa	55 MPa
Allowable electric field (kV/cm)	2.5	3.5	4.5	2.5	3.0	4.0
d_{15} (pC/N)	2100	1750	1600	3000	2750	2600
Maximum strain	0.5‰	0.6‰	0.7‰	0.7‰	0.8‰	1.0‰

TABLE III. Transverse dielectric loss factor tanδ_E of PIN-PMN-PT crystals upon various uniaxial stresses and ac electric fields.

	"1T" crystal			"2R" crystal		
	0 MPa	15 MPa	25 MPa	0 MPa	30 MPa	55 MPa
1 kV/cm	1.50%	0.95%	0.86%	0.20%	0.20%	0.12%
2 kV/cm	4.10%	2.10%	1.40%	1.40%	0.80%	0.70%
3 kV/cm	10.3%	3.37%	3.20%	6.70%	2.10%	1.70%
4 kV/cm	>50.0%	6.50%	4.20%	20.0%	7.00%	3.90%

The work supported by the National Nature Science Foundation of China (Grant Nos. 51102193 and 51002116), National Basic Research Program of China (973 Program) under Grant No. 2009CB623306 and International Science and Technology Cooperation Program of China under Grant No. 2010DFR50480.

[1]S.-E. Park and T. R. Shrout, J. Appl. Phys. **82**, 1804 (1997).
[2]B. Noheda, Curr. Opin. Solid State Mater. Sci. **6**, 27 (2002).
[3]M. Davis, J. Electroceram. **19**, 23 (2007).
[4]D. Damjanovic, IEEE Trans. Ultrason. Ferroelectr. Freq. Control **56**, 1574 (2009).
[5]F. Li, S. Zhang, Z. Xu, X. Wei, J. Luo, and T. R. Shrout, J. Appl. Phys. **108**, 034106 (2010).
[6]G. S. Xu, K. Chen, and D. F. Yang, Appl. Phys. Lett. **90**, 032901 (2007).
[7]S. J. Zhang, J. Luo, W. Hackenberger, and T. R. Shrout, J. Appl. Phys. **104**, 064106 (2008).
[8]S. J. Zhang and F. Li, J. Appl. Phys. **111**, 031301 (2012).
[9]S -F. Liu, W. Ren, B. K. Mukherjee, S. J. Zhang, T. R. Shrout, P. W. Rehrig, and W. S. Hackenberger, Appl. Phys. Lett. **83**, 2886 (2003).
[10]F. Li, S. J. Zhang, Z. Xu, X. Wei, and T. R. Shrout, Adv. Funct. Mater. **21**, 2118 (2011).
[11]R. Zhang, B. Jiang, and W. Cao, Appl. Phys. Lett. **82**, 3737 (2003).
[12]S. J. Zhang, L. Lebrun, S. F. Liu, S. Rhee, C. A. Randall, and T. R. Shrout, Jpn. J. Appl. Phys. **41**, 1099 (2002).
[13]S. J. Zhang, F. Li, J. Luo, R. Xia, W. Hackenberger, and T. R. Shourt, Appl. Phys. Lett. **97**, 132903 (2010).
[14]P. Han, W. Yan, J. Tian, X. Huang, and H. Pan, Appl. Phys. Lett. **86**, 052902 (2005).
[15]S. J. Zhang, F. Li, W. H. Jiang, J. Luo, R. J. Meyer, Jr., W. W. Cao, and T. R. Shrout, Appl. Phys. Lett. **98**, 182903 (2011)
[16]F. Li, S. J. Zhang, Z. Xu, X. Wei, J. Luo, and T. R. Shrout, Appl. Phys. Lett. **97**, 252903 (2010).
[17]S. J. Zhang, G. Liu, W. H. Jiang, J. Luo, W. W. Cao, and T. R. Shrout, J. Appl. Phys. **110**, 064108 (2011).
[18]S. J. Zhang, F. Li, J. Luo, R. Xia, W. Hackenberger, and T. R. Shourt, IEEE Trans. Ultrason. Ferroelectr. Freq. Control. **28**, 274–280 (2011).
[19]Y. H. Xu, Y. J. Feng, and N. Zhang, J. Am. Ceram. Soc. **94**, 3863 (2011).
[20]M. Budimir, D. Damjanovic, and N. Setter, Phys. Rev. B **72**, 064107 (2005).
[21]E. Burcsu, G. Ravichandran, and K. Bhattacharya, Appl. Phys. Lett. **77**, 1698 (2000).
[22]J. J. Gao, Z. Xu, F. Li, C. H. Zhang, Y. Liu, G. M. Liu, and H. L. He, J. Appl. Phys. **109**, 114111 (2011).
[23]A. Amin, M. J. Haun, B. Badger, Jr., H. A. McKinstry, and L. E. Cross, Ferroelectrics **65**, 107 (1985).
[24]K. Uchino and S. Hirose, IEEE Trans. Ultrason. Ferroelectr. Freq. Control **48**, 307 (2001).