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Orientation dependence of electromechanical properties of relaxor based ferroelectric single crystals

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Abstract The orientation dependence of electromechanical properties of relaxor based ferroelectric single crystals $Pb(Zn_{1/3}Nb_{2/3})O_3$–(6–7)%$PbTiO_3$ and $Pb(Mg_{1/3}Nb_{2/3})O_3$–33%$PbTiO_3$ has been calculated by coordinate transformation. Different from previous studies, the optimum cutting orientations have been predicted in terms of their piezoelectric responses in the corresponding crystal planes. The calculation results indicated that the anisotropic piezoelectric effects of $[001]_c$ and $[011]_c$ poled multi-domain crystals mainly come from the intrinsic contribution. However, the strong dielectric anisotropy of $[001]_c$ poled multi-domain crystals mainly comes from extrinsic domain and domain wall contributions. For $[011]_c$ poled multi-domain crystals, the intrinsic orientation effect enhances the dielectric anisotropy.

Introduction

Relaxor based ferroelectric single crystals, such as $(1-x)Pb(Zn_{1/3}Nb_{2/3})O_3$–$xPbTiO_3$ (PZN–xPT) and $(1-x)Pb(Mg_{1/3}Nb_{2/3})O_3$–$xPbTiO_3$ (PMN–xPT), have attracted considerable attention in recent years because of their extraordinary large piezoelectric coefficient d_{33} (>2000 pC/N) and electromechanical coupling coefficient k_{33} (>0.90) when being poled along $[001]_c$ of cubic coordinates [1–3]. However, some experimental observations have demonstrated that the enhanced electromechanical performance is not limited to $[001]_c$ poled crystals. Viehland et al. have found high-piezoelectric constants up to 5000–6000 pC/N and electromechanical coupling coefficients up to 0.95 in $[011]_c$ oriented PZN–xPT crystals [4]. It was reported that ultrahigh transverse piezoelectric property of PMN–30%PT crystal occurs in crystals with the polarization (thickness) and vibration (length) directions along $[110]_c$ and $[001]_c$, respectively [5]. This phenomenon was further verified by Wang et al. who reported full matrix electromechanical coefficients of $[011]_c$ PMN–29%PT single crystals [6].

Using orientation to enhance certain functional properties has been recognized as an important method since the electromechanical properties of many single crystals are highly anisotropic. The orientation dependence of piezoelectric properties of PMN–33%PT and PZN–9%PT single crystals has been described theoretically by using the Euler's angles [7–11]. Such representation may be difficult to visualize and often causes confusion. For example, in Ref. [10, 11], the rotation plane is the $(1\bar{1}0)$ crystal plane, but it is more appropriate to use the $(0\bar{1}1)$ plane, which contains the $[011]_c$ direction so that the theoretical values can be compared to the experimental observations in the $[011]_c$ poled crystals. Here, through coordinate transformation [12], we directly define the crystal plane of rotation based on the actual coordinates used in the experiments, which makes it easier to visualize and less confusing when making comparison with actual experimental observations. Previous efforts were mostly focused on the d_{33} value by using coordinate transformation method [13]. Attempts on the calculation of orientation dependence of transverse or shear piezoelectric properties need to consider the rotation effect around the transformed z-axis, so that the calculated results can be properly compared with experimentally measured results. For example, to compare the calculated

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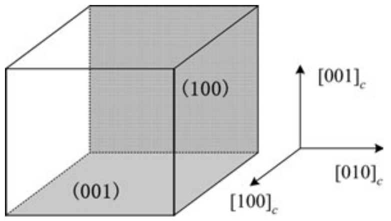

Fig. 1 Crystal planes (100) and (001) in cubic coordinates

results with the experimental data of $[001]_c$ poled PMN–33%PT single crystal, the orientation dependence of piezoelectric coefficient d_{31} could be calculated either on the crystal plane (100) or (001), as illustrated in Fig. 1.

Davis and Damjanovic et al. have calculated the orientation dependence of d_{31} for PMN–33%PT on the crystal plane (001) in terms of Euler's angles [9]. We will discuss the orientation dependence of piezoelectric and dielectric properties of PZN–(6–7)%PT and PMN–33%PT single crystals on the crystal plane (100) by using coordinate transformation method. In addition, the orientation dependence of properties of these single crystals on the crystal plane $(0\bar{1}1)$ will also be calculated. The calculated results are then compared with available multi-domain experimental data in the same coordinate system, which can provide more direct guidance to the crystal growers and more credible evidence to the origin of the piezoelectric and dielectric properties anisotropy of multi-domain crystals.

Calculation method

Coordinate transformation method has been extensively applied to calculate crystal properties and investigate the effect of crystal orientation on piezoelectric response of ferroelectric crystals [13–16]. Generally speaking, the macroscopic electromechanical properties of ferroelectric crystals mainly come from two effects: one is the intrinsic orientation effect, i.e., the effect of crystal anisotropy of a single-domain single crystal; the other is the extrinsic contribution from domains and domain walls, i.e., the effect of averaging over the crystallographically allowed orientations of ferroelectric domains and domain walls in a multi-domain single crystal. For example, for a single-domain PMN–33%PT crystal [17], its intrinsic orientation plays a crucial role in determining their macroscopic electromechanical properties. While for a multi-domain PMN–33%PT crystal [3], both intrinsic orientation and extrinsic domain structures determine the crystal properties. If the properties of single-domain PMN–33%PT crystal were used as input data, the properties in different orientations could be effectively calculated by coordinate transformation method. Therefore, if the properties of single-domain and multi-domain PMN–33%PT crystals were obtained in the same coordinate system, it would be easier to quantify the contributions from the intrinsic orientation and extrinsic domain structure contribution to the functional properties of multi-domain crystals.

The original single-domain data used in this work are the piezoelectric and dielectric coefficients of single-domain PMN–33%PT [17] and PZN–(6–7)%PT [18] crystals in their intrinsic coordinate system: $x \leftrightarrow [1\bar{1}0]_c$, $y \leftrightarrow [11\bar{2}]_c$, and $z \leftrightarrow [111]_c$. To describe the orientation dependence of properties in single-domain crystals on the (100) crystal plane, a coordinate system: $x \leftrightarrow [100]_c$, $y \leftrightarrow [01\bar{1}]_c$, and $z \leftrightarrow [011]_c$ is treated as a transitional coordinate system. The properties of single-domain crystal in the intrinsic coordinate system are firstly transformed to the transitional coordinate system, then, rotated counterclockwise around the fixed axis $x \leftrightarrow [100]_c$ to obtain the orientation dependence of piezoelectric properties in terms of the rotation angle (θ). The angle between the transitional $z \leftrightarrow [011]_c$ and transformed z-axis is defined as θ. When the rotation angle θ equals 45°, the corresponding coordinate system becomes: $x \leftrightarrow [100]_c$, $y \leftrightarrow [010]_c$, and $z \leftrightarrow [001]_c$, which matches the coordinate system used in the experimental measurement for $[001]_c$ poled multi-domain crystals [3, 19].

Similarly, the orientation dependence of properties of single-domain PZN–(6–7)%PT and PMN–33%PT crystals on the $(0\bar{1}1)$ crystal plane can be calculated. A transitional coordinate system is: $x \leftrightarrow [0\bar{1}1]_c$, $y \leftrightarrow [2\bar{1}\bar{1}]_c$, and $z \leftrightarrow [111]_c$, then, the properties are rotated counterclockwise around $x \leftrightarrow [0\bar{1}1]_c$. The angle between the original $z \leftrightarrow [111]_c$ and transformed z-axis is defined as θ. When the rotation angle θ equals 35.3°, the corresponding coordinate system is: $x \leftrightarrow [0\bar{1}1]_c$, $y \leftrightarrow [100]_c$, and $z \leftrightarrow [011]_c$, which will match the coordinate system used in the experiment for $[011]_c$ poled multi-domain crystal [6, 20].

Results and discussions

Orientation dependence of properties of PZN–(6–7)%PT crystal on the (100) crystal plane

For single-domain PZN–(6–7)%PT crystal, the orientation dependence of piezoelectric coefficients d_{31} and d_{33} on the crystal plane (100) is shown in Figs. 2 and 3, respectively. Dashed line represents negative values, while solid line represents positive values of corresponding quantities. As shown in Fig. 2, when the rotation angle θ is equal to 0° or 180°, the absolute value of d_{31} reaches its maximum (2119 pC/N). While the absolute maximum value of d_{33} (3053 pC/N) appears at four rotation angles ($\theta = 49.8°$, 229.8°, 130.2°, and 310.2°), as shown in Fig. 3. The

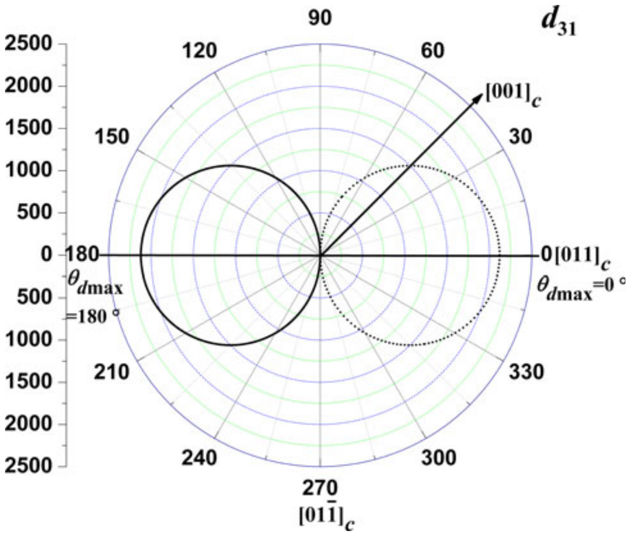

Fig. 2 Orientation dependence of piezoelectric coefficient d_{31} (pC/N) of PZN–(6–7)%PT single crystal on the crystal plane (100)

Table 1 Piezoelectric constants d_{3j} (pC/N) ($j = 1$ or 3) and dielectric permittivities ε^T_{mn} (ε_0) ($m = n = 1$ or 3) calculated for single-domain crystals and measured in $[001]_c$ poled multi-domain crystals [3, 19] in the coordinate system: $x \leftrightarrow [100]_c$, $y \leftrightarrow [010]_c$, and $z \leftrightarrow [001]_c$

	Measured properties of PZN–7%PT [19]	Rotated properties of PZN–(6–7)%PT
d_{31}	−1204	−1499
d_{33}	2455	3010
ε^T_{11}	3000	7567
ε^T_{33}	5622	7567
	Measured properties of PMN–33%PT [3]	Rotated properties of PMN–33%PT
d_{31}	−1330	−1152
d_{33}	2820	2309
ε^T_{11}	1600	2847
ε^T_{33}	8200	2847

calculated properties of single-domain PZN–(6–7)%PT crystal in the coordinate system: $x \leftrightarrow [100]_c$, $y \leftrightarrow [010]_c$, and $z \leftrightarrow [001]_c$ are given in Table 1. Since there had been no reports in the literature on the complete set of electromechanical coefficients of $[001]_c$ poled multi-domain PZN–(6–7)%PT crystal up to date, the measured properties of multi-domain PZN–7%PT crystal [19] were listed in Table 1 for comparison with the calculated results in the same coordinate system.

It can be seen from Table 1 that quantitative matching between the calculation and the experimental measurement is not perfect for neither piezoelectric coefficients nor dielectric permittivities. These deviations are mainly caused by two factors: one is that the calculations only take into account the crystal intrinsic orientation effect, excluding the extrinsic domain structure contributions to the electromechanical properties of the multi-domain single crystals; the other is due to experimental errors.

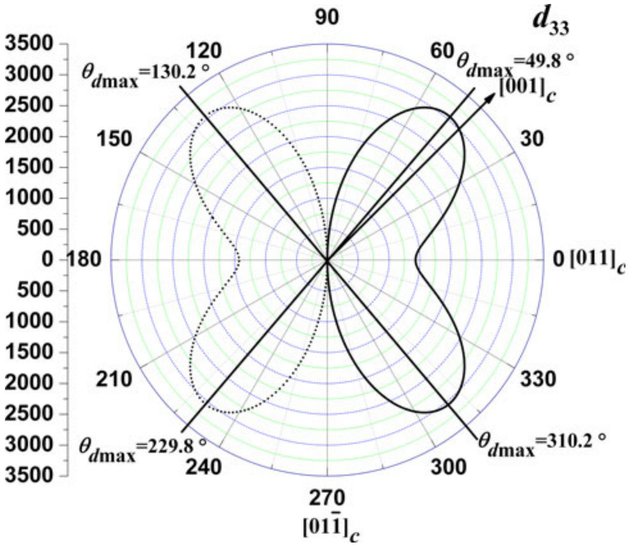

Fig. 3 Orientation dependence of piezoelectric coefficient d_{33} (pC/N) of PZN–(6–7)%PT single crystal on the crystal plane (100)

The calculated results show that ε^T_{11} and ε^T_{33} of single-domain PZN–(6–7)%PT crystal are equal in the coordinate system: $x \leftrightarrow [100]_c$, $y \leftrightarrow [010]_c$, and $z \leftrightarrow [001]_c$, as shown in Table 1. Thus, the extrinsic contribution from the domain structures is dominant in determining the observed strong anisotropy in the dielectric properties of the multi-domain crystal [19]. In addition, it has been pointed out that the domain structure strongly affects the dielectric properties of tetragonal PZN–12%PT crystal [14] and rhombohedral PMN–33%PT crystals [15]. If considering the domain wall contributions, it will reduce ε^T_{11} but enhance ε^T_{33} [15], so that the dielectric anisotropy occurs in the $[001]_c$ poled multi-domain PZN–7%PT crystal [19].

In terms of experimental errors, some are caused by chemical compositional fluctuations, others are from the measurements of electromechanical properties of single-domain PZN–(6–7)%PT crystal. These experimental errors will be transferred and amplified in the calculations. If a quantity $y = f(x_1, x_2, x_3, \ldots)$ is related to some quantities $x_1, x_2, x_3, \ldots$ with errors Δx_1, Δx_2, $\Delta x_3, \ldots$ the error of y can be expressed as [21]

$$\Delta y = \sum_i \left|\frac{\partial f}{\partial x_i}\right| \Delta x_i. \tag{1}$$

The expression of d_{31} for single-domain PZN–(6–7)%PT crystal in the coordinate system: $x \leftrightarrow [100]_c$, $y \leftrightarrow [010]_c$, and $z \leftrightarrow [001]_c$ is

$$d_{31} = 0.2721d_{22}^{111} + 0.3848d_{31}^{111} + 0.1924d_{33}^{111} - 0.1924d_{15}^{111}, \qquad (2)$$

where d_{ij}^{111} ($i = 1, 2, 3, j = 1, 2, 3, 5$) are the piezoelectric coefficients of single-domain PZN–(6–7)%PT crystal in the coordinate system: $x \leftrightarrow [1\bar{1}0]_c$, $y \leftrightarrow [11\bar{2}]_c$, and $z \leftrightarrow [111]_c$[18].

From Eq. 1, the relative error of d_{31} can be described as

$$\frac{\Delta d_{31}}{d_{31}} = 0.2307\frac{\Delta d_{22}^{111}}{d_{22}^{111}} + 0.0090\frac{\Delta d_{31}^{111}}{d_{31}^{111}} + 0.0119\frac{\Delta d_{33}^{111}}{d_{33}^{111}} + 0.7705\frac{\Delta d_{15}^{111}}{d_{15}^{111}}. \qquad (3)$$

If the chemical composition difference between the crystals used in Refs. [18] and [19] were ignored, the relative error of calculated d_{31} would mainly come from the measurements of piezoelectric coefficients d_{22}^{111} and d_{15}^{111} in the single-domain PZN–(6–7)%PT crystal.

The expression of d_{33} using single-domain PZN–(6–7)%PT crystal data in the coordinate system: $x \leftrightarrow [100]_c$, $y \leftrightarrow [010]_c$, and $z \leftrightarrow [001]_c$ is

$$d_{33} = -0.5443d_{22}^{111} + 0.3847d_{31}^{111} + 0.1942d_{33}^{111} + 0.3847d_{15}^{111}. \qquad (4)$$

The relative error $\Delta d_{33}/d_{33}$ of d_{33} can be expressed as

$$\frac{\Delta d_{33}}{d_{33}} = 0.2315\frac{\Delta d_{22}^{111}}{d_{22}^{111}} + 0.0045\frac{\Delta d_{31}^{111}}{d_{31}^{111}} + 0.0060\frac{\Delta d_{33}^{111}}{d_{33}^{111}} + 0.7671\frac{\Delta d_{15}^{111}}{d_{15}^{111}}. \qquad (5)$$

Obviously, the relative error $\Delta d_{33}/d_{33}$ also mainly comes from d_{22}^{111} and d_{15}^{111} measurements in the single-domain crystal.

The error analysis can also be performed on calculated dielectric permittivities ε_{11}^{T} and ε_{33}^{T} for single-domain PZN–(6–7)%PT crystal listed in Table 1. The dielectric permittivities ε_{11}^{T} and ε_{33}^{T} have the same expression as Eq. 6 in the coordinate system: $x \leftrightarrow [100]_c$, $y \leftrightarrow [010]_c$, and $z \leftrightarrow [001]_c$:

$$\varepsilon_{11}^{T} = 0.6667\varepsilon_{11}^{111} + 0.3333\varepsilon_{33}^{111}, \qquad (6)$$

where ε_{ij}^{111} ($i = j = 1,3$) are the dielectric permittivities of single-domain PZN–(6–7)%PT crystal in coordinate system: $x \leftrightarrow [1\bar{1}0]_c$, $y \leftrightarrow [11\bar{2}]_c$, and $z \leftrightarrow [111]_c$.

The relative error $\Delta\varepsilon_{11}^{T}/\varepsilon_{11}^{T}$ is

$$\frac{\Delta\varepsilon_{11}^{T}}{\varepsilon_{11}^{T}} = 0.9692\frac{\Delta\varepsilon_{11}^{111}}{\varepsilon_{11}^{111}} + 0.0308\frac{\Delta\varepsilon_{33}^{111}}{\varepsilon_{33}^{111}}. \qquad (7)$$

If the composition variation were ignored, the relative error of calculated ε_{11}^{T} would mainly come from the measurement of ε_{11}^{111} in single-domain PZN–(6–7)%PT crystal. According to Eq. 6, the measured dielectric permittivity ε_{11}^{111} may be too large.

Orientation dependence of properties of PMN–33%PT crystal on the crystal plane (100)

The orientation dependence of piezoelectric coefficients d_{31} and d_{33} on the crystal plane (100) of PMN–33%PT single crystal was calculated. The absolute maximum value of d_{31} (1629 pC/N) occurs at $\theta = 0°$ and 180°, and the absolute maximum value of d_{33} (2360 pC/N) appears at four rotation angles ($\theta = 50.7°$, 129.3°, 230.7°, and 309.3°). These locations are near the {100} family orientations, so that $[001]_c$ poled rhombohedral PZN–xPT and PMN–xPT single crystals have very large piezoelectric coefficients. The measured multi-domain properties reported in Ref. [3] together with the single-domain properties calculated through coordinate rotation to the coordinate system: $x \leftrightarrow [100]_c$, $y \leftrightarrow [010]_c$, and $z \leftrightarrow [001]_c$, are listed in Table 1 for comparison. It can be seen that the rotated piezoelectric coefficients d_{31} (≈ -1152 pC/N, $\theta = 45.0°$) and d_{33} (≈ 2309 pC/N, $\theta = 45.0°$) are about 87 and 82% of the corresponding crystal properties measured in the multi-domain state. It demonstrated that for $[001]_c$ poled PMN–33%PT single crystals, the intrinsic orientation effect plays a significant role in determining its piezoelectric coefficients d_{31} and d_{33}.

The calculated rotated dielectric permittivities ε_{11}^{T} and ε_{33}^{T} of single-domain PMN–33%PT crystal are equal as shown in Table 1, which means that the intrinsic orientation effect contributes equally to ε_{11}^{T} and ε_{33}^{T} for $[001]_c$ poled PMN–33%PT crystal, the strong anisotropy in the dielectric properties observed in multi-domain PMN–33%PT crystal [3] should be attributed to the extrinsic domain and domain wall contributions. For the PMN–33%PT single crystal, the domain wall contribution reduced ε_{11}^{T} but enhanced ε_{33}^{T} [15].

Orientation dependence of properties of PZN–(6–7)%PT crystal on the crystal plane $(0\bar{1}1)$

Figures 4 and 5 show the orientation effects of d_{31} and d_{33} of PZN–(6–7)%PT single crystal on the crystal plane $(0\bar{1}1)$, respectively. The optimal values of piezoelectric coefficients $d_{31} \approx \pm 1280$ pC/N ($\theta = 91.6°$ or 271.6°) and $d_{33} \approx \pm 3080$ pC/N ($\theta = 119.7°$ or 299.7°) have been obtained. When the rotation angle (θ) equals 35.3°, the coordinate system becomes: $x \leftrightarrow [0\bar{1}1]_c$, $y \leftrightarrow [100]_c$, and $z \leftrightarrow [011]_c$, corresponding to the one used for $[011]_c$ poled crystal in the multi-domain state [20]. The rotated properties of single-domain PZN–(6–7)%PT crystal and its multi-domain properties measured [20] are given in Table 2. Error analysis showed that the main error origins of the calculated piezoelectric coefficients come from d_{22}^{111} and d_{15}^{111} measurements in the single-domain PZN–(6–7)%PT crystal. The errors of calculated dielectric

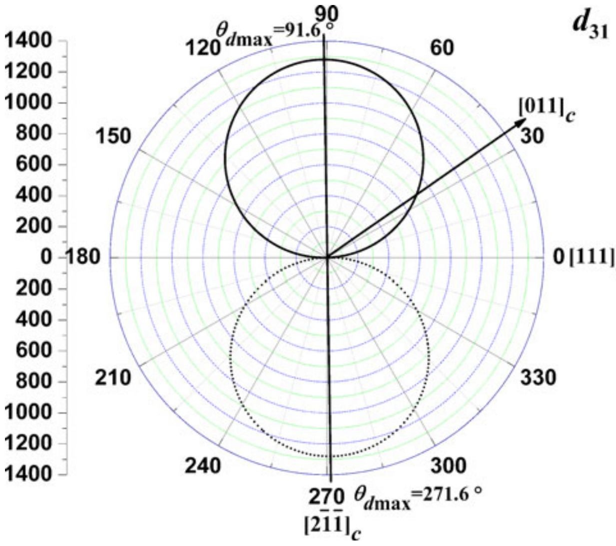

Fig. 4 Orientation dependence of piezoelectric coefficient d_{31} (pC/N) of PZN–(6–7)%PT single crystal on the crystal plane $(0\bar{1}1)$

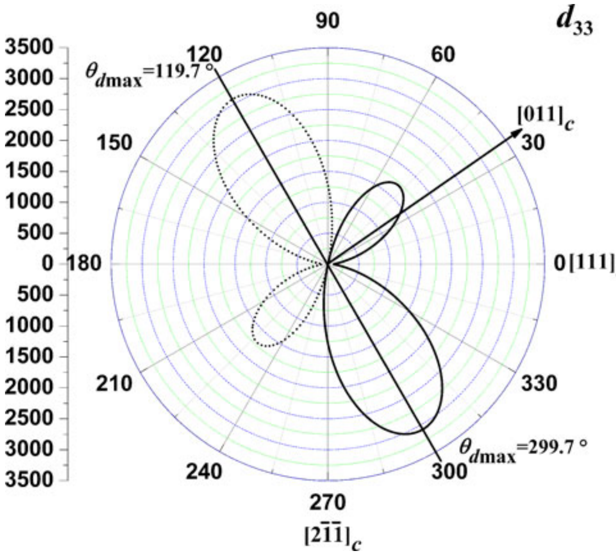

Fig. 5 Orientation dependence of piezoelectric coefficient d_{33} (pC/N) of PZN–(6–7)%PT single crystal on the crystal plane $(0\bar{1}1)$

permittivities mainly come from ε_{11}^{111} measurement in the single-domain crystal. These error origins of piezoelectric and dielectric properties calculated for $[011]_c$ poled PZN–(6–7)%PT are actually the same as those for $[001]_c$ poled crystals.

Orientation dependence of properties of PMN–33%PT crystal on the crystal plane $(0\bar{1}1)$

Table 2 Piezoelectric constants d_{3j} (pC/N) (j = 1 or 3) and dielectric permittivities ε_{mn}^{T} (ε_0) ($m = n$ = 1 or 3) calculated for single-domain crystals and measured in $[011]_c$ poled multi-domain crystals [6, 20] in the coordinate system: $x \leftrightarrow [0\bar{1}1]_c$, $y \leftrightarrow [100]_c$, and $z \leftrightarrow [011]_c$

	Measured properties of PZN–7%PT [20]	Rotated properties of PZN–(6–7)%PT
d_{31}	478	711
d_{33}	1150	1429
ε_{11}^{T}	8240	11000
ε_{33}^{T}	3180	4139
	Measured properties of PMN–29%PT [6]	Rotated properties of PMN–33%PT
d_{31}	610	701
d_{33}	1020	938
ε_{11}^{T}	3564	3950
ε_{33}^{T}	4033	1745

The orientation dependence of d_{31} and d_{33} of single-domain PMN–33%PT crystal on the crystal plane $(0\bar{1}1)$ has been calculated. The absolute maximum values of piezoelectric coefficients d_{31} (1343 pC/N) and d_{33} (2411 pC/N) occur at $\theta = 93.8°$ (or 273.8°) and $\theta = 117.1°$ (or 297.1°), respectively. The calculated piezoelectric and dielectric properties of single-domain PMN–33%PT crystal in the coordinate system: $x \leftrightarrow [0\bar{1}1]_c$, $y \leftrightarrow [100]_c$, and $z \leftrightarrow [011]_c$ are given in Table 2. Since no complete set of electromechanical coefficients of $[011]_c$ poled multi-domain PMN–33%PT crystal had been measured so far, the measured properties of $[011]_c$ poled PMN–29%PT crystal [6] are listed in Table 2 for comparison. The calculation results showed that the crystal intrinsic orientation effect can indeed enhance the anisotropy of dielectric and piezoelectric properties of $[011]_c$ poled PMN–33%PT crystals.

Conclusions

The orientation dependence of electromechanical properties of single-domain PZN–(6–7)%PT and PMN–33%PT crystals on the crystal planes (100) and $(0\bar{1}1)$ has been calculated by coordinate transformation method. The optimum cutting orientations were predicted on these two crystal planes in terms of piezoelectric coefficients d_{31} and d_{33}. Compared with available experimental data in the literature, our calculated results show that the crystal intrinsic orientation effect is the primary contributor to the observed anisotropic piezoelectric properties of $[001]_c$ and $[011]_c$ poled crystals. On the other hand, the intrinsic orientation effect makes equal contribution to the dielectric properties ε_{11}^{T} and ε_{33}^{T} of the crystals in the coordinate system: $x \leftrightarrow [100]_c$, $y \leftrightarrow [010]_c$, and $z \leftrightarrow [001]_c$, which suggests that the domain structures play a very important role in determining the dielectric anisotropy of $[001]_c$ poled single crystals. While for the $[011]_c$ poled crystals, crystal intrinsic orientation effect makes different contributions to dielectric properties ε_{11}^T and ε_{33}^T, which enhances the anisotropy of the dielectric properties.

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