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Optimization of electrooptic and piezoelectric coupling effects in tetragonal relaxor-PT ferroelectric single crystals

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Abstract

The electrooptic and piezoelectric coupling effects in tetragonal relaxor-based ferroelectric 0.62Pb(Mg_{1/3}Nb_{2/3})O₃-0.38PbTiO₃ (PMN-0.38PT) and 0.88Pb(Zn_{1/3}Nb_{2/3})O₃-0.12PbTiO₃ (PZN-0.12PT) single-domain crystals have been analyzed by the coordinate transformation. The orientation dependence of the electrooptic and half-wave voltage was calculated based on the full sets of refractive indices, electrooptic and piezoelectric coefficients. The optimum orientation cuts for achieving the best electrooptic coefficient and half-wave voltage were found. The lowset half-wave voltage is only 76 V for the PMN-0.38PT single-domain crystal. Compared to commonly used electrooptic crystal LiNbO₃, tetragonal relaxor-PT ferroelectric single-domain crystals are much superior for optical modulation applications because of their much higher linear electrooptic coefficients and substantially lower half-wave voltages when the piezoelectric strain influence is considered.

Keywords

Electrooptic; piezoelectric; coupling effect; PMN-PT; PZN-PT

1. Introduction

Ferroelectric crystals have been widely used in optical devices of multi-functions, such as electrooptic (EO) modulators, holographic storage systems, and laser frequency multipliers [1–3]. The LiNbO₃ crystal is one of the best known ferroelectrics used in electrooptic modulations. However, the electrooptic and piezoelectric coefficients of LiNbO₃ crystal are low ($r_{33}=32.2$ pm/V and $d_{33}=6$ pC/N) [4]. It was found that the tetragonal relaxor-PT single-

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domain crystals have much better electrooptic and piezoelectric properties than LiNbO₃ crystals. For example, the EO coefficient r_{33} of [001] poled tetragonal PMN-0.38PT and PZN-0.12PT single-domain crystals are 70 pm/V and 134 pm/V, respectively, which are much larger than that of LiNbO₃ crystal ($r_{33}=32.2$ pm/V) [5,6]. Larger EO coefficients correspond to lower half-wave voltage, which would enable EO modulators to have smaller physical dimensions and operated at a lower voltage [7,8].

Recently, it was found that the large piezoelectric strain constants d_{31} and d_{33} have significant influence to the half-wave voltage of relaxor-PT electrooptic modulators [9], due to the strong coupling between the electrooptic modulation and the inverse piezoelectric effect. Hence, the orientation optimization of electrooptic and piezoelectric coupling effects in tetragonal relaxor-PT single crystals is important for the design of EO modulators, which has not aroused the attention of researchers up to date.

In this work, a systematic analysis was performed to study the spatial anisotropies of linear electrooptic and piezoelectric effects of [001] poled tetragonal ferroelectric PMN-0.38PT and PZN-0.12PT single crystals. Our results showed that the optimum electrooptic and piezoelectric coupling effects do not appear along their principal axes, and the electric field induced strain has significant influence to the performance of electrooptic modulators due to dimensional changes caused by the strong inverse piezoelectric effect.

2. Computational method

Both linear electrooptic and piezoelectric coefficients are third-rank tensors. The [001] poled PMN-0.38PT and PZN-0.12PT single crystals belong to the tetragonal phase with $4mm$ point group symmetry at room temperature [10,11]. Their full sets of refractive indices n_o and n_e [6], linear electrooptic coefficients r_{13} , r_{33} , and r_{51} [12,13], and piezoelectric strain coefficients d_{31} , d_{33} , and d_{15} [14,15] were listed in Table 1 and Table 2, which are much larger than that of LiNbO₃ crystal ($r_{33}=32.2$ pm/V and $d_{33}=6$ pC/N) [4]. It should be noted that the refractive indices and unclamped linear EO coefficients listed in Table 1 were measured under the illumination of 632.8 nm light. The linear EO effect describes the induced change n_{ij} in the refractive index due to an applied electric field E

$$\Delta n_{ij} = -\frac{1}{2}n_{ij}^3 \sum_{k=1}^3 r_{ijk} E_k \quad (1)$$

where n_{ij} is the refractive index and r_{ijk} is the linear EO coefficient.

Three dimensional analysis of spatial anisotropy of the linear EO and piezoelectric effects is performed based on the linear coordinate transformation. The linear EO tensor component r'_{unh} in the rotated coordinate system is related to the tensor component r_{pcq} in the original coordinate system by the following relation [16]

$$r'_{unh} = a_{up} a_{nc} a_{hq} r_{pcq} \quad (2)$$

where a_{up} , a_{nc} , a_{hq} are the components of the transformation matrix. Similarly, the transformation of piezoelectric strain coefficient by changing the coordinate system is represented by the following equation [17]

$$d'_{ijk} = a_{il}a_{jm}a_{kn}d_{lmn} \quad (3)$$

The linear EO tensor $\{r_{ijk}\}$ is of the same form as the converse piezoelectric tensor d_{ijk} because of the permutation symmetry of the first two indices. Therefore, the linear EO and piezoelectric strain tensors in Voigt notation can also be written as $\{r_{\lambda j}\}$ and $\{r_{j\lambda}\}$, with $\lambda=1-6$, $j=1-3$.

3. Results and discussion

Fig. 1 shows the spatial anisotropy of the linear EO coefficient r'_{33} of PMN-0.38PT single crystal based on its full set of linear EO coefficient data (r_{13} , r_{33} , and r_{51}). The maximum value of r'_{33} (453 pm/V) is on the [100]-[001] plane with an angle $\theta = 53^\circ$ from the [001] axis, as shown in Fig. 2, which is 6.5 times as large as that in its principal crystallographic axis ($r_{33}=70$ pm/V). Fig. 3 shows the spatial anisotropy of the linear EO coefficient r'_{33} of LiNbO₃ single crystal based on its full set of linear EO coefficient data ($r_{13}=10$ pm/V, $r_{33}=32.2$ pm/V, $r_{22}=6.81$ pm/V and $r_{51}=32$ pm/V) for comparison [4]. The maximum value of r'_{33} is 39.9 pm/V, which is only 1.2 times as large as that in its principal crystallographic axis ($r_{33}=32.2$ pm/V). The maximum value of r'_{33} for PMN-0.38PT is 11.4 times as large as that of LiNbO₃. Fig. 4 shows the spatial anisotropy of the piezoelectric strain coefficient d'_{32} based on the full set of piezoelectric strain coefficient data (d_{31} , d_{33} , and d_{15}) for PMN-0.38PT. The maximum value of d'_{32} is along the original [001] axis on the [100]-[001] plane. Our calculations show that the optimized orientations of the linear EO and piezoelectric effect are very different.

Half-wave voltage V_π is an important parameter for EO modulators, which has different values for different operation modes. Fig. 5(a) shows a typical transverse EO phase modulation system [9]. A polarizer is inserted in front of the crystal to produce a polarized light and the external electric field E is in the z-direction ([001]-axis to the crystal). The incident light is along the y-axis ([010]-axis to the crystal). For the phase modulator, the polarization direction of the polarizer is parallel to the induced electro-optic principal axis and the allowed directions of polarization are the x- and z-directions, so that r_{13} and r_{33} components of the EO tensor can be used. With this configuration, the polarization state of the incident light parallel to one of the principal axes will not change but the phase will change with applied electric field. Due to the inverse piezoelectric effect, the phase variation corresponding to r'_{33} component before and after the crystal being subjected to an electric field in the new rotated coordinate system is [9]

$$\varphi'_{33} = -\frac{2\pi}{\lambda} \left[\left(-\frac{1}{2}n_e^3 r'_{33} + n_e' d'_{32} \right) \frac{L}{d} V \right] \quad (4)$$

where L is the length along the optical path, d is the electrode spacing, d'_{32} is the transverse piezoelectric constant in the new rotated coordinate system, and V is the applied voltage.

For the transverse intensity modulation, the EO crystal is placed between a linear polarizer and a linear analyzer, as shown in Fig. 5(b). The polarizer direction is 45° with respect to the z -axis of the coordinate system and the polarization direction of the analyzer is perpendicular to the polarizer. The polarization of the input laser beam will be decomposed into x - and z - components. The phase difference between the two light polarization components considering transverse piezoelectric strain is expressed by [9]

$$\Gamma^{T'} = -\frac{2\pi}{\lambda} \left\{ \left[-\frac{1}{2} n'_e r'_c + (n'_e - n'_o) d'_{32} \right] \frac{L}{d} V \right\} \quad (5)$$

where $r'_c = r'_{33} - (n'_o/n'_e)^3 r'_{13}$ is the effective electro-optic coefficient in the new rotated coordinate system.

For phase modulation, the half-wave voltage V_π is the voltage required for inducing a phase change of $+\pi$ or $-\pi$, while for the intensity modulator, V_π is the voltage required to change the operation condition from minimum transmission to maximum transmission. Fig. 6 shows the orientation dependence of $V_{\pi 33}^{T'}$ (based on r'_{33}) and $V_{\pi c}^{T'}$ (based on r'_c) for [001] poled PMN-0.38PT single crystal on the [100]-[001] plane. The theta (θ) is the angle away from the [001] axis in the counterclockwise direction. As listed in Table 3, we can see that the minimum values of $V_{\pi 33}^{T'}$ and $V_{\pi c}^{T'}$ are 76 V when $\theta=53^\circ$ and 81 V when $\theta=54^\circ$, which are much lower than that in the original coordinate system, in which $V_{\pi 33}^T$ and $V_{\pi c}^T$ are 338 V and 813 V, respectively. While for the EO modulation based on r'_{13} , the optimization of the $V_{\pi 13}^T$ appears in the original [001] axis, i.e., $\theta=0^\circ$, because the optimum values of r'_{13} and d'_{33} are along the original [001] axis. However, the value of $V_{\pi 13}^T$ is very large (580 V).

The spatial anisotropies of electrooptic, piezoelectric, and half-wave voltage of [001] poled PZN-0.12PT single crystal display similar characteristics and the optimized cuts of electrooptic and piezoelectric coupling effect were listed in Table 1–3. The minimum values of $V_{\pi 33}^{T'}$ and $V_{\pi c}^{T'}$ are 94 V when $\theta=51^\circ$ and 102 V when $\theta=53^\circ$, which are much lower than that in the original coordinate system, in which $V_{\pi 33}^T$ and $V_{\pi c}^T$ are 210 V and 345 V for PZN-0.12PT single crystal.

Moreover, it must be mentioned that the half-wave voltage for the transverse modulation can be decreased by increasing the aspect ratio L/d . For example, the optimized half-wave voltages $V_{\pi 33}^{T'}$ and $V_{\pi c}^{T'}$ of PMN-0.38PT single crystal are only 7.6 V with 53° -cut and 8.1 V with 54° -cut, respectively, when $L=10d$. In addition, the [001] poled tetragonal relaxor-PT single crystals with stable single-domain structure have good compositional uniformity, good thermal stability, and high optical transmittance [18–21]. The transmittance of [001] poled PMN-0.38PT single-domain crystal with 1 mm thickness is 63% at the wavelength of 632.8 nm [22]. The reflectance caused by the two faces of the crystal can be calculated by Fresnel formula to be 31% based on the refractive index of 2.5 [23]. So the scattering loss is

only 6%. If the two faces of this crystal are coated with antireflective film, the transmittance can reach more than 90%.

4. Summary and conclusions

When EO crystals are in mechanically stress-free state and operate at low frequency electric field, there is obvious coupling between the electrooptic and piezoelectric effects. The large piezoelectric constants have significant influence to the half-wave voltage of electrooptic modulators. We have predicted the optimum values of electrooptic and piezoelectric coupling coefficients using coordinate transformation of the full matrix data. For [001] poled PMN-0.38PT and PZN-0.12PT single crystals, the optimum cut for the EO modulation applications is in the [100]-[001] plane with an angle of 51–54° from the [001] axis.

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There exists strong electrooptic and piezoelectric coupling effects in PMN-PT.
The optimum cuts for achieving the best electrooptic coefficient were found.
The lowset half-wave voltage is only 76 V for the PMN-0.38PT single-domain crystal.
Relaxor-PT ferroelectric crystals are much superior for optical application.

Fig. 1.

The spatial anisotropy of the linear EO coefficient r'_{33} of [001] poled PMN-0.38PT single crystal.

Fig. 2.

The orientation dependence of r'_{33} on the [100]-[001] plane for [001] poled PMN-0.38PT.

Fig. 3.
The spatial anisotropy of the linear EO coefficient r'_{33} of LiNbO₃ single crystal.

Fig. 4.
The spatial anisotropy of the piezoelectric coefficient d'_{32} of [001] poled PMN-0.38PT single crystal.

Fig. 5. Schematic drawing of (a) transverse electro-optic phase modulation and (c) intensity modulation.

Fig. 6.

The orientation dependence of $V''_{\pi 33}$ and $V''_{\pi c}$ on the [100]-[001] plane for [001] poled PMN-0.38PT single crystal.

Table 1

Refractive indices and linear EO coefficients of ferroelectric PMN-0.38PT and PZN-0.12PT single-domain crystals.

Crystals	n_o	n_e	r_{13} ($\text{pm}\cdot\text{V}^{-1}$)	r_{33} ($\text{pm}\cdot\text{V}^{-1}$)	r_{51} ($\text{pm}\cdot\text{V}^{-1}$)	r_c ($\text{pm}\cdot\text{V}^{-1}$)	r'_{33} ($\text{pm}\cdot\text{V}^{-1}$)	r'_{13} ($\text{pm}\cdot\text{V}^{-1}$)	r'_c ($\text{pm}\cdot\text{V}^{-1}$)
PMN-0.38PT	2.616	2.600	25	70	558	44	453 ($\theta=53^\circ$)	15 ($\theta=53^\circ$)	438 ($\theta=54^\circ$)
PZN-0.12PT	2.57	2.46	7	134	462	131	387 ($\theta=51^\circ$)	4 ($\theta=51^\circ$)	382 ($\theta=52^\circ$)

Table 2

Piezoelectric strain constants of ferroelectric PMN-0.38PT and PZN-0.12PT single-domain crystals.

Crystals	d_{31} (pC·N ⁻¹)	d_{33} (pC·N ⁻¹)	d_{15} (pC·N ⁻¹)	d'_{33} (pC·N ⁻¹)	d'_{32} (pC·N ⁻¹)
PMN-0.38PT	-123	300	380	164 ($\theta=53^\circ$)	-58 ($\theta=53^\circ$)
PZN-0.12PT	-207	541	653	304 ($\theta=51^\circ$)	-94 ($\theta=51^\circ$)

Table 3

The calculated half-wave voltages with considering converse piezoelectric effect with $L=d$ for ferroelectric PMN-0.38PT and PZN-0.12PT single-domain crystals.

Crystals	$V_{\pi 33}^T$ (V)	$V_{\pi c}^T$ (V)	$V_{\pi 13}^T$ (V)	$V_{\pi 33}^{T'}$ (V)	$V_{\pi c}^{T'}$ (V)
PMN-0.38PT	338	813	580	76 ($\theta=53^\circ$)	81 ($\theta=54^\circ$)
PZN-0.12PT	210	345	535	94 ($\theta=51^\circ$)	102 ($\theta=53^\circ$)