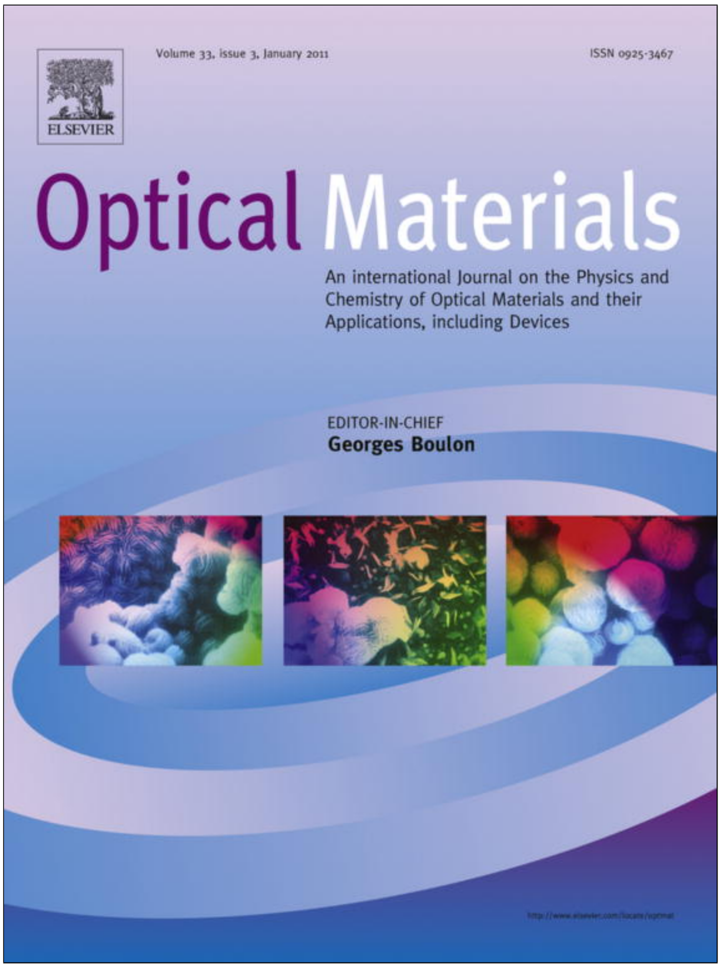
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Reduction of electro-optic half-wave voltage of $0.93Pb(Zn_{1/3}Nb_{2/3})O_3–0.07PbTiO_3$ single crystal through large piezoelectric strain

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A B S T R A C T

The influence of converse piezoelectric effect on the electro-optic coefficient of single domain relaxor-based $0.93Pb(Zn_{1/3}Nb_{2/3})O_3–0.07PbTiO_3$ (PZN-0.07PT) has been quantified under ambient conditions. It was found that the large piezoelectric constants d_{31} and d_{33} have significant influence to the half-wave voltage of electro-optic modulators. For single domain PZN-0.07PT crystal, $V^T_{\pi 13}$ is reduced by a factor of 8 and $V^L_{\pi 13}$ can be decreased by more than an order of magnitude due to the large piezoelectric effect. Compared to commonly used electro-optic crystal $LiNbO_3$ and $BaTiO_3$, PZN-*x*PT single crystal is much superior for optic phase modulation applications because they have much higher linear electro-optic coefficients and much lower half-wave voltage when piezoelectric strain influence is considered.

1. Introduction

Electro-optic (E-O) crystals have many applications, such as spatial light modulators [1], optical switches [2] and volume holographic memories [3]. Most of bulk electro-optic modulation devices operate on the basis of transverse and/or longitudinal electro-optic effects. In optical modulations, low operating voltage is preferred. Generally speaking, larger aspect ratio and higher linear electro-optic coefficients are the two key factors for decreasing the half-wave voltage of the modulator.

Ferroelectric single crystals are commonly used as optical functional material because of their excellent electro-optic, photorefractive, and nonlinear optical properties. In recent years, there is an increasing interest in relaxor-based ferroelectric single crystals, $(1-x)Pb(Zn_{1/3}Nb_{2/3})O_3–xPbTiO_3$ (PZN-*x*PT) and $(1-x)Pb(Mg_{1/3}Nb_{2/3})O_3–xPbTiO_3$ (PMN-*x*PT), which were found to possess both very high piezoelectric constants [4–6] and very large linear electro-optic coefficients [7–11]. For example, the electro-optic coefficients of tetragonal PZN-0.12PT [7] and PMN-0.38PT [9] single crystals poled along $[0\,0\,1]_c$ direction are: r_{33} = 134 and 70 pm/V, respectively, which are much larger than that of the well-known E-O crystal $LiNbO_3$ (r_{33} = 30.8 pm/V) [12]. Larger electro-optic coefficients correspond to lower half-wave voltage, which would enable E-O modulators to have smaller physical dimensions and being operated at lower voltage.

PZN-0.07PT single crystal, which is slightly away from morphotropic phase boundary (MPB) composition, has compositional uniformity, good thermal stability, and can be poled in single domain state [13,14]. The transmittance of $[1\,1\,1]_c$ poled PZN-0.07PT single domain single crystal with 1 mm thickness is 0.63 at the wavelength of 632.8 nm [15]. The reflectance caused by the two faces of the crystal can be calculated by Fresnel formula to be 0.31 based on the refractive index of 2.5. So the scattering loss is only 6%. If the two faces of this crystal are coated with anti-reflective film, the transmittance can reach more than 90%.

In this work, we have characterized the piezoelectric strain constants and linear electro-optic coefficients of $LiNbO_3$ (LN) and $[1\,1\,1]_c$-poled $0.93Pb(Zn_{1/3}Nb_{2/3})O_3–0.07PbTiO_3$ (PZN-0.07PT) single crystals at room temperature. Then, a systematic analysis was performed to study the influence of piezoelectric strain constants d_{31} and d_{33} on the electro-optic phase modulation and intensity modulation using $LiNbO_3$, and single domain PZN-0.07PT. The results are compared to that of $BaTiO_3$ single crystals [16]. Our results showed that the field induced strain has significant influence to electro-optic modulators due to dimensional changes caused by the piezoelectric effects.

2. Experimental

The PZN-0.07PT single crystals used in this work were grown by the high temperature flux technique [13]. Following the convention of relaxor ferroelectric crystals, we define $[1\,\bar{1}\,0]_c$, $[1\,1\,\bar{2}]_c$, and $[1\,1\,1]_c$ of the pseudo-cubic directions as the *a*, *b* and *c* axes,

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respectively. Gold electrodes were deposited onto the $[1\,1\,1]_c$ and $[\bar{1}\,\bar{1}\,\bar{1}]_c$ surfaces through ion sputtering. The samples were poled along $[1\,1\,1]_c$ in silicone oil at room temperature by applying an electric field of 4 kV/cm for 30 min. The $[1\,1\,\bar{2}]_c$ faces were polished to optical quality for optical measurements. A k_{31} piezoelectric vibrator was prepared for measuring the transverse piezoelectric constant d_{31}.The $LiNbO_3$ and PZN-0.07PT single crystals are all in rhombohedral phase with *3m* point group symmetry at room temperature. When applying an electric field E_3 along the *c*-axis, the refractive index changes due to linear electro-optic effect are

$$\Delta n_o = -\frac{1}{2} n_o^3 r_{13} E_3 \tag{1a}$$

$$\Delta n_e = -\frac{1}{2} n_e^3 r_{33} E_3 \tag{1b}$$

where n_o and n_e denote the ordinary and extraordinary refractive indices, respectively; r_{13} ($r_{13} = r_{23}$) and r_{33} are the linear electro-optic coefficients under stress free condition. The piezoelectric coefficients have no influence to the clamped electro-optical coefficients.

The refractive indices were measured by the Brewster's angle method and the linear electro-optic coefficients were measured at low frequency (1 kHz) by the modified Mach–Zehnder interferometric method. Details on the measurements can be found in Ref. [17]. The light source is a 632.8 nm He–Ne laser. For the electro-optic coefficient measurements, the applied electric field along the *c*-axis will cause a change in crystal dimension along the optical pathway due to the converse piezoelectric effect, which is given by

$$\Delta L = d_{31} E_3 L = d_{31} L \frac{V}{d} \tag{2}$$

where L is the length along the optical path, d is the electrode spacing, d_{31} ($d_{31} = d_{32}$) is the transverse piezoelectric constant, and V is the applied voltage. Hence, the measured apparent linear electro-optic coefficients r^*_{13} and r^*_{33} are:

$$r^*_{13} = r_{13} - 2\frac{(n_o - 1)}{n_o^3} d_{31} \tag{3a}$$

$$r^*_{33} = r_{33} - 2\frac{(n_e - 1)}{n_e^3} d_{31} \tag{3b}$$

The intrinsic values of electro-optic coefficients r_{13} and r_{33} can be derived from Eq. (3a). During electro-optic coefficient measurements, the polarization direction of the laser beam is perpendicular to the *c*-axis for measuring r^*_{13}, and parallel to the *c*-axis for measuring r^*_{33}. The electro-optic measurement system was calibrated by testing a $LiNbO_3$ crystal for which the electro-optic properties are well documented in the literature. All measured optical parameters of $LiNbO_3$ single crystal in our work agree well with data in the literature [12,18], which validated our measurement setup.The transverse piezoelectric strain constant d_{31} of PZN-0.07PT single crystals was determined by the resonance method using a standard k_{31} piezoelectric vibrator [19]. The longitudinal piezoelectric strain constant d_{33} was directly measured by quasi-static method using a ZJ-2 piezo d_{33} meter.

3. Results and discussion

The measured material constants of $LiNbO_3$ and PZN-0.07PT single crystals are listed in Table 1. From the measured results, we can see that the PZN-0.07PT has comparable piezoelectric and electro-optic properties to other compositions of relaxor-based ferroelectric PZN-PT and PMN-PT single crystals [7,9], which are much larger than that of $LiNbO_3$ single crystal [12].Half-wave voltage V_π is an important parameter for electro-optic modulators. There are different V_π for phase modulators and intensity modulators, and also for different mode of operations. Fig. 1(a) shows a typical transverse electro-optic phase modulation system. The *c*-axis of the E-O crystal is along the *z*-direction of the coordinate system. 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. The incident light is along the *y*-axis (*b*-axis to the crystal). For the phase modulator, we use r_{13} and r_{33} components of the E-O effects so that the polarization direction of the polarizer must be parallel to the induced electro-optic principal axis and the allowed directions of polarization are the *x*- and *z*- directions. 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 piezoelectric effect, the phase variations corresponding to r_{13} and r_{33} ccomponents before and after the crystal being subjected to an electric field are:

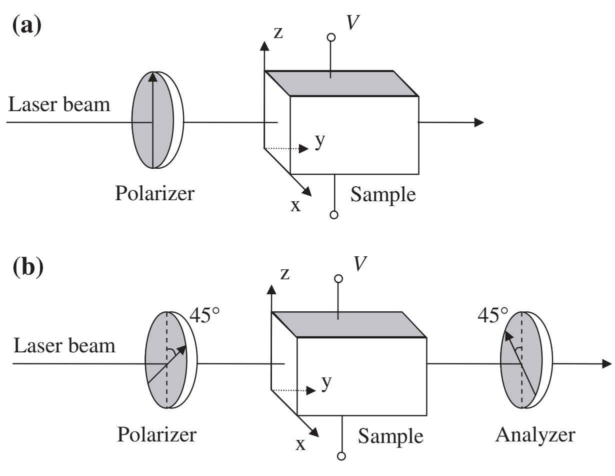

Fig. 1. Schematic drawing of typical transverse electro-optic phase (a) and intensity (b) modulations.

Table 1
Measured refractive indices, apparent electro-optic coefficients, piezoelectric strain constants, and calculated half-wave voltages with (*) and without (no *) considering converse piezoelectric effect with $L = d$ for $LiNbO_3$, $BaTiO_3$, and PZN-0.07PT single-domain ferroelectric single crystals.

Single Crystals	n_o	n_e	r^*_{13} (pm V^{-1})	r^*_{33} (pm V^{-1})	$-d_{31}$ (pC N^{-1})	d_{33} (pC N^{-1})		
$LiNbO_3$	2.288	2.203	10	31	1	9		
$BaTiO_3$ [16]	2.412	2.360	11.2	113	32.5	90		
PZN-0.07PT	2.466	2.488	23	90	97	180		
	$V^{T}_{\pi13}$ (kV)	$V^{T^*}_{\pi13}$ (kV)	$V^{T}_{\pi33}$ (kV)	$V^{T^*}_{\pi33}$ (kV)	$V^{T}_{\pi c}$ (kV)	$V^{T^*}_{\pi c}$ kV)	$V^{L}_{\pi13}$ (kV)	$V^{L^*}_{\pi13}$ (kV)
$LiNbO_3$	5.399	5.196	1.923	1.898	2.987	2.990	5.399	8.325
$BaTiO_3$	6.465	2.485	0.443	0.400	0.475	0.476	6.465	1.882
PZN-0.07PT	10.460	1.174	0.577	0.400	0.610	0.608	10.460	0.765

$$\begin{aligned}\varphi_{13}^{T^*} &= -\frac{2\pi}{\lambda}\Delta(n_o L) \\ &= -\frac{2\pi}{\lambda}(\Delta n_o L + n_o \Delta L) \\ &= -\frac{2\pi}{\lambda}\left[\left(-\frac{1}{2}n_o^3 r_{13} + n_o d_{31}\right)\frac{L}{d}V\right] \end{aligned} \quad (4a)$$

$$\begin{aligned}\varphi_{33}^{T^*} &= -\frac{2\pi}{\lambda}\Delta(n_e L) \\ &= -\frac{2\pi}{\lambda}(\Delta n_e L + n_e \Delta L) \\ &= -\frac{2\pi}{\lambda}\left[\left(-\frac{1}{2}n_e^3 r_{33} + n_e d_{31}\right)\frac{L}{d}V\right] \end{aligned} \quad (4b)$$

For a longitudinal phase modulator, the c-axis of the E-O crystal will be along the y-axis of the coordinate system, which is also the incident light direction. In this configuration, the effective electro-optic component is only r_{13}. Considering the longitudinal piezoelectric strain, the phase variation caused by the applied electric voltage is:

$$\varphi_{13}^{L^*} = -\frac{2\pi}{\lambda}\left[\left(-\frac{1}{2}n_o^3 r_{13} + n_o d_{33}\right)V\right] \quad (5)$$

For the transverse intensity modulation, the E-O crystal is placed between a linear polarizer and a linear analyzer. 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 configuration is shown in Fig. 1(b). The polarization of the input laser beam will be decomposed into x- and z-components. The optical transmittance T of the modulation system is given by [20]

$$T = \frac{I}{I_0} = T_0 \sin^2\left(\frac{\Gamma^{T^*}}{2}\right) \quad (6)$$

where I_0 and I are the input and output light intensities, respectively; T_0 is the transmission factor and Γ^{T^*} is the phase difference between the two light polarization components considering transverse piezoelectric strain.

When an electric voltage is applied on the crystal along the z-axis, there will be a field-induced birefringence in the (x, z) plane

$$\Delta n_{xz} = -\frac{1}{2}n_e^3 r_c \frac{V}{d} \quad (7)$$

where r_c is the effective electro-optic coefficient. It can be calculated from the measured values of n_o, n_e, r_{13}, and r_{33} by the following formula:

$$r_c = r_{33} - \left(\frac{n_o}{n_e}\right)^3 r_{13} \quad (8)$$

When the transverse piezoelectric strain is considered, the phase difference between the two light polarization components may be expressed by

$$\Gamma^{T^*} = -\frac{2\pi}{\lambda}\left\{\left[-\frac{1}{2}n_e^3 r_c + (n_e - n_o)d_{31}\right]\frac{L}{d}V\right\} \quad (9)$$

In general, there will be different half-wave voltages in different modulation types. For phase modulation, the half-wave voltage V_π is the voltage required for inducing a phase change by $+\pi$ or $-\pi$, while for the intensity modulator, V_π is the voltage required to change the operation condition from minimum transmission to maximum transmission. The calculated half-wave voltages V_π (without considering converse piezoelectric effect) and V_π^* (considering converse piezoelectric effect) of $LiNbO_3$, $BaTiO_3$, and PZN-0.07PT single crystals with the dimension of $L = d$ in different modulation types are also listed in Table 1 (Note: the actual dimension is not important here but only the aspect ratio of the E-O crystal). We can see that V_π and V_π^* differ substantially for all ferroelectric single crystals listed in Table 1, particularly for phase modulation based on r_{13}.

In the transverse phase modulation, the half-wave voltages considering transverse piezoelectric strain become much lower for all ferroelectric single crystals listed in Table 1. According to Eq. (4a), the transverse piezoelectric effect always helps to reduce the half-wave voltage because $d_{31} < 0$. While for the transverse intensity modulation, the difference between V_π and V_π^* is not significant because the value ($n_e - n_o$) in Eq. (5) is very small for ferroelectric single crystals listed here. 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 half-wave voltage $V_{\pi 33}^{T^*}$ of PZN-0.07PT single crystal is only 40 V when $L = 10d$.

For the longitudinal phase modulation, the longitudinal piezoelectric strain constant d_{33} has different influence on the half-wave voltage for different ferroelectric single crystals. It actually increased the half-wave voltage for $LiNbO_3$, but has positive influence for $BaTiO_3$ and PZN-0.07PTsingle crystals. This is because $LiNbO_3$ single crystal has much smaller piezoelectric strain constant d_{33} than the other two.

4. Conclusions

When electro-optic crystals are in mechanically stress-free state and operate at low frequency or dc electric field, the dimensional change of the piezoelectric crystal caused by the applied electric field will make significant contribution to the E-O modulation properties. Based on our experimental and computational analyses, the converse piezoelectric effect plays an important role in the actual electro-optic devices. The reduction for $V_{\pi 13}^{T}$ is by a factor of 8 to reach a value of 1.174, and for $V_{\pi 13}^{L}$ the reduction is more than an order of magnitude to reach a value of only 0.74. Our results showed that the PZN-0.07PT single crystal is much superior as an E-O phase modulator material due to its larger piezoelectric strain constants, higher electro-optic coefficients, and especially the very lower half-wave voltage induced by the piezoelectric effect. To achieve the same phase modulation, the length of the modulator made of PZN-0.07PT single crystal is only about 1/5 of the modulator made of $LiNbO_3$ single crystal with the same lateral dimensions of and under 40 V operating voltage.

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