

Field-induced light deflection in lithium niobate and lithium tantalate: the possible configurations

L. Guilbert

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Abstract Field-induced light deflection by ferroelectric domain walls in lithium niobate (LN) and lithium tantalate (LT) is theoretically studied. The phenomenon can occur not only when both the incident wave vector k and the electric field E are parallel to the z -axis—as demonstrated by experiments made so far—but also when E and/or k are perpendicular to z . In particular, for E parallel to x and k parallel to y , the deflection phenomenon is predicted to have the same characteristics as in triclinic or monoclinic ferroelastics: the large deflection angle is related to the natural birefringence, whereas the deflection amplitude is proportional to the small tilt angle of the neutral lines, which is here induced by the electric field. In periodic domain structures, interference between deflected waves occurs, and the deflected intensity is expected to be largely enhanced when the Bragg condition is satisfied. This transverse configuration is thus specially attractive to characterize periodically-poled crystals.

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1 Introduction

Domain patterning in ferroelectrics (FE) such as lithium niobate (LN) or lithium tantalate (LT) is of rising interest. In particular, one-dimensional periodic domain structures (PPLN, PPLT) are used for quasi-phase-matching in nonlinear optical applications, and two-dimensional arrays of

tubular domains are promising for photonic bandgap crystals. Non-destructive methods are required to characterize domain structures during or after the patterning process. Among them, optical methods have been somewhat neglected for many years, because in FE crystals such as LN or LT domain structures are in principle invisible owing to symmetry.

However, when an electric field is applied on a multidomain FE crystal, the electro-optical (EO) effect modifies the refractive indices differently in the different orientation states, and thus can give an optical contrast to the domain structure. Recently, the light-deflection phenomenon has been evidenced in LN [1] and LT [2] crystals submitted to high electric fields. Several light spots appear in the far field when a laser beam crosses the domain structure. During the experiments the authors have directed both the electric field and the incident beam along the polar axis. This deflection phenomenon is due to refraction at domain walls [3]. To some extent, it is similar to the well-known deflection in ferroelastics (FEL) [4–10], except that, in the specific configuration used in Refs. [1, 2] for LN and LT, domain walls are not mirror planes for the optical indices of neighbor domains. For this reason, deflection occurs from positive domains (spontaneous polarization P_s parallel to E) to negative domains (P_s antiparallel to E), but the reverse process is impossible at grazing incidence in this configuration. One spectacular consequence of this asymmetry has been observed in LT [2]: deflection by triangular domains does not give six-fold but three-fold patterns, except when total reflection occurs inside the domains.

The aim of the present paper is to predict what the characteristics of the field-induced light deflection could be in other configurations. The nature of the field-induced optical discontinuities at domain walls depends on the directions of

L. Guilbert (✉)
Laboratoire Matériaux Optiques Photonique et Systèmes,
University Paul Verlaine, CNRS UMR 7132, Supélec 2,
rue Edouard Belin, 57070 Metz, France
e-mail: guilbert@metz.supelec.fr

the applied field and the incident wave vector, and their magnitude depends on the active EO coefficient. Four cases of practical interest with respect to $R3m$ crystals (LN, LT) will be reviewed.

2 Possible configurations

Domain walls will be supposed to be parallel to the plane (yz), as commonly observed in LN and LT. The incident wave vector at grazing incidence on domain walls can thus be parallel to either y or z . The electric field can be applied along z , y , or x . The four cases of main interest are gathered in Table 1.

(a) $E // z, k // z$

This is the experimental configuration chosen by Müller et al. in Refs. [1, 2]. The crystal remains uniaxial and the active EO coefficient is r_{13} . As theoretically demonstrated in Ref. [3], the deflection angle is almost proportional to the square root of the applied field, and the phenomenon is

asymmetrical: deflection occurs only from positive domain to negative domain (Fig. 1a). The ordinary beam (polarized along y) and the extraordinary beam (polarized perpendicular to y) are deflected at similar angles, given by

$$\sin \alpha_o \approx n_o^2 \times (2r_{13}E_z)^{1/2} \tag{1}$$

$$\sin \alpha_e \approx n_o n_e \times (2r_{13}E_z)^{1/2} \tag{2}$$

Since $n_o \approx n_e$, it is probably difficult to separate the two polarizations in the deflected beams. It can thus be said that deflection patterns are only weakly sensitive to incident polarization in this configuration. The deflected intensity is related to the domain wall density in the section of the incident beam but the relationship is not straightforward at grazing incidence.

(b) $E // z, k // y$

This configuration has been already utilized for the fabrication of EO beam deflectors [11, 12]. It does not differ much from the previous one: the phenomenon is also asymmetrical (Fig. 1b) and the deflection angle is again proportional to the

Table 1 Expected characteristics of field-induced deflection in LN or LT for different configurations of applied field and incident light

	$E // z, k // z$	$E // z, k // y$	$E // x$ or $y, k // z$	$E // x, k // y$
Active EO coefficients	r_{13}	r_{13}, r_{33}	$r_{61} (= -r_{22})$	r_{51}
Sensitivity to polarization	no	yes	no	yes
Symmetry at grazing incidence	no	no	yes	yes
Deflection angle (sinus)	$n^2 \times (2r_{13}E)^{1/2}$	$n_i^2 \times (2r_{i3}E)^{1/2}$	$n_o n_e \times (2r_{22}E)^{1/2}$	$(n_o^2 - n_e^2)^{1/2}$

Fig. 1 Construction of the deflected beams on the slowness curves, for incident beam parallel to z (left) and to y (right). The electric field is parallel to z in both cases. The upper view shows the illuminated face of the crystal, with the index ellipsoid differently modified by the electric field in the two neighbor domains

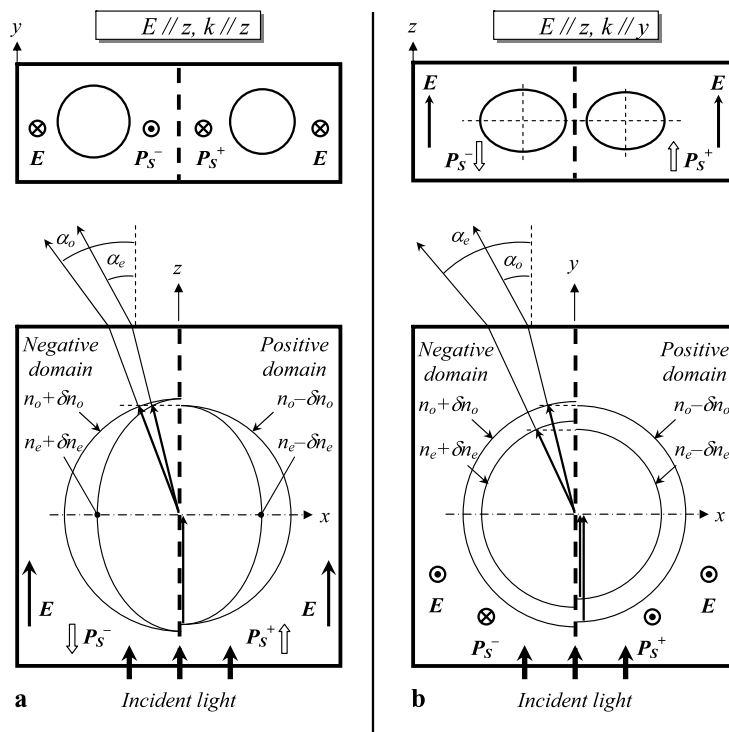
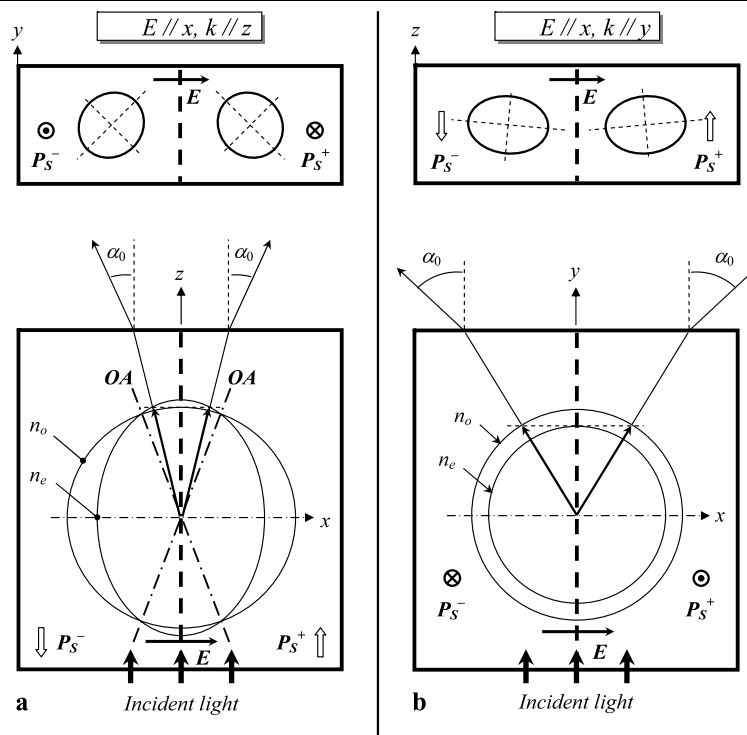


Fig. 2 Construction of the deflected beams on the slowness curves, for incident beam parallel to z (left) and to y (right). The electric field is parallel to x in both cases. The upper view shows the illuminated face of the crystal, with the index ellipsoid differently modified by the electric field in the two neighbor domains (the neutral axes, shown by dash lines, are electrically induced in the first case)



square root of the applied field in very good approximation. However, the active EO coefficient is r_{33} when the incident beam is polarized along z , whereas it is r_{13} for incident polarization along x . The corresponding deflection angles are given by

$$\sin \alpha_o \approx n_o^2 \times (2r_{13}E_z)^{1/2} \tag{3}$$

$$\sin \alpha_e \approx n_e^2 \times (2r_{33}E_z)^{1/2} \tag{4}$$

Note that both deflected beams are ordinary (subscript e only means that the corresponding wave velocity is c/n_e). Contrary to the previous case, the beams can be separated easily, because r_{33} is much larger than r_{13} (about 3 times in LN, 6 times in LT). Deflection patterns should thus be highly sensitive to incident polarization. This configuration is suitable rather for layered domain structures (PPLN or PPLT). In the case of tubular domains parallel to z , complicated deflection patterns are expected, owing to multiple refraction processes.

(c) $E // x$ (or y), $k // z$

When the electric field is applied along x and the incident wave vector parallel to z , the active EO coefficient is $r_{61}(= -r_{22})$ and the crystal becomes biaxial. The index ellipsoid is strained in the (xy) plane: initially isotropic with respect to the polarization of the incident light, the crystal becomes birefringent, with neutral lines at 45° from x and y . The denomination ‘positive’ or ‘negative’ for the domains becomes arbitrary. The slow axis of positive domains coincides with the fast axis of negative domains and vice versa.

At grazing incidence on a single domain wall, the deflection consists of two symmetrical beams (Fig. 2a), called A and A' according to Tsukamoto’s notation [4]. Contrary to the first configuration, triangular domains of LT should always give here six-fold deflection patterns. The deflection angle at grazing incidence is again almost proportional to the square root of the applied field:

$$\sin \alpha_o \approx n_o n_e \times (2r_{22}E_y)^{1/2} \tag{5}$$

In this configuration, field-induced deflection in LN or LT is similar to natural deflection in orthorhombic FEL crystals such as gallium molybdate (GMO) or potassium dihydrogen phosphate (KDP) when the incident beam is close to the c -axis. If the LN or LT sample is rotated in the (zx) plane, new deflected beams (called B and B') should appear as soon as the incidence angle exceeds α_0 . This feature can be easily understood by reversing the light path. Increasing incidence further, the A -beam and the B -beam should merge on the direct beam, and then split again, exchanging their polarizations. This ‘crossing point’ at incidence angle i_c (reported for the first time by Tsukamoto et al. in GMO [5]) corresponds to propagation along one of the two optical axes (OA), which are here splitted symmetrically by the electric field at small angle from the polar axis. Figure 3 helps to understand the deflection phenomenon at small incidences ($\alpha_0 < i < i_c$): the lightwave changes velocity and thus direction alternatively from domain to domain but keeps its polarization almost unchanged. The division of the wave amplitude at each domain wall is very weak. Deflection proceeds

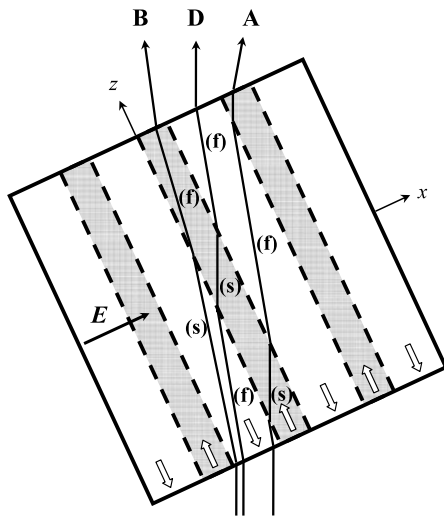


Fig. 3 Deflection at small incidence in the third configuration ($E//x$, k close to z), when the incidence angle is larger than α_0 (deflection angle at grazing incidence) but smaller than i_c (the so-called “crossing” incidence at which A , B and D merge all together). Symbols (s) and (f) refer to slow and fast waves respectively. Polarization directions of A and B beams are crossed to one another, at 45° from y . The birefringence and the incidence angle have been exaggerated for clarity

here essentially by division of the wavefront: the beam is deviated only if it enters the crystal by a negative domain and gets out by a positive domain or vice versa. Consequently, in the case of PPLN or PPLT crystals the deflected intensity at small incidence mainly depends on the duty cycle ρ of the domain structure, that is, the width of positive domains divided by the full period of the domain structure. The deflected intensity at small incidence should be maximal for $\rho = 50\%$. This configuration could thus be used to scan the duty cycle along a PPLN or PPLT crystal. But its interest looks somewhat limited because it cannot give any information on the period of the domain structure, contrary to the configuration that will be studied in subsection (d).

Note that applying the electric field along y instead of x has a similar effect on the ellipsoid in the (xy) plane, except that the field-induced neutral lines appear along x and y . Hence the configuration $\{E//y, k//z\}$ does not bring about anything more compared to $\{E//x, k//z\}$.

(d) $E//x, k//y$

In this configuration the EO coefficient r_{51} induces a tilting of the index ellipsoid in the (zx) plane. Since this tilting is symmetrical on each side of the domain wall, deflection is also symmetrical (Fig. 2b). The mutual tilt angle 2ϕ is proportional to the electric field:

$$\tan 2\phi = \frac{r_{51} E_x}{1/n_o^2 - 1/n_e^2} \quad (6)$$

The situation is the same as in low-symmetry FEL's (e.g., Rochelle salt [4], RbHSeO₄ [6, 7] BaCl₂ [7], APFA [8]).

The deflection angle α_0 of A -beams at grazing incidence is related to the natural birefringence:

$$\sin^2 \alpha_0 = |n_o^2 - n_e^2| \quad (7)$$

This angle is independent of the applied field, contrary to the previous cases. At 633 nm, (7) gives $\alpha_0 = 38.4^\circ$ in LN, 7.6° in LT. Varying the incidence in the (xy) plane, the deflection angle α relative to the y -axis increases with the incidence angle i , as follows [7]:

$$\sin^2 \alpha \approx |n_o^2 - n_e^2| + \sin^2 i \quad (8)$$

In LN and LT, since one has $n_o > n_e$, A -beams are polarized in the incident plane. B -beams appear for $i > \alpha_0$ and are polarized along z . At small incidence on one domain wall, according to Tsukamoto's approximation [4], the amplitude of the deflected wave relatively to the incident one is

$$a \approx \sin 2\phi \approx \frac{\bar{n}^3 r_{51} E_x}{2\Delta n} (\ll 1), \quad (9)$$

with $\bar{n}^3 = 2n_o^2 n_e^2 / (n_o + n_e)$, $\Delta n = n_o - n_e$. The deflected intensity is thus proportional to E^2 . For instance, in LN under field $E_x = 100$ kV/cm, (9) gives a deflection intensity ratio $a^2 \approx 4.5 \times 10^{-4}$ at 633 nm, using the following data: $n_o = 2.286$, $n_e = 2.200$, $r_{51} = 32.6$ pm/V [13]. Field-induced deflection by an isolated domain wall in this configuration should thus be weak but nevertheless detectable.

Contrary to the case previously studied in subsection (c), deflection proceeds here by amplitude division at domain walls. If the incident beam crosses several domain walls, the parallel-deflected waves interfere and add their complex amplitudes. Therefore, the total deflected intensity depends on the number of domain walls and on the phase difference between the deflected waves. The latter itself depends on the distance between domain walls, the incidence angle and the wavelength. Interferential deflection has been experimentally shown in glycine phosphite [9] and has been theoretically treated in Ref. [10]. In a periodic domain structure, diffraction and deflection occur simultaneously. Deflection can be either strong or weak, depending on whether the deflected waves interfere constructively or destructively. Hence, by rotating a periodic domain structure in a monochromatic beam, one expects to observe a maximum of deflection each time the deflected spot crosses a diffracted spot—in other words, each time the Bragg condition is fulfilled at the deflection angle. The sharpness of the enhancement will reflect the periodicity of the domain structure. The transverse configuration with E parallel to the x -axis and k in the (yz) plane is thus specially interesting to check periodically-poled crystals after domain switching.

3 Conclusion

This theoretical work opens the way to further experimental investigation of field-induced light deflection. The characteristics of the phenomenon in LN and LT are expected to depend on the directions of the electric field E and of the incident beam. For E parallel to the y -axis and the incident beam in the (zx) plane, deflection should have the same characteristics as natural deflection in high-symmetry FEL's (GMO, KDP). For E parallel to the x -axis and the incident beam in the (yx) plane, deflection should have the same characteristics as natural deflection in low-symmetry FEL's (RHSe, APFA). The latter configuration seems specially attracting to characterize PPLN or PPLT crystals.

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