A simple method for photoconductivity measurement in lithium niobate

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Supplementary information

The application of our method to lithium niobate is based on electro-optic modulation in the so-called $r_{22}$ configuration: the light propagates along the $z$-axis and the electric field is applied on the $y$-axis. This configuration is favorable because the influence of thermo-optic effects can be neglected. However, due to the photogalvanic effect (sometimes dubbed also as Bulk Photovoltaic Effect) there exist a photogalvanic (PG) current $J_{PG}$ induced by the light in the crystal bulk. Thus a PG field builds up in the illuminated area. As our method is based on the time dependence of the internal fields, as probed by the electro-optic refractive index change, it is important to assess whether the internal PG field can affect the results of our measurement.

The photogalvanic current in lithium niobate can be written as:

$$J_j = \left( \beta_{jkl}^S + i \beta_{jkl}^A \right) E_k E_l^*$$

where $E_{k(l)}$ are the optical field polarization components and $\beta_{jkl}^S$ and $\beta_{jkl}^A$ are the symmetric and anti-symmetric parts of the PG tensor, respectively. For the three components of the current, this can be rewritten explicitly taking into account the form of the PG tensor in the Voigt notation for lithium niobate:

$$J_x = \beta_{22}^S \left( E_x E_y^* + E_y E_x^* \right) + \beta_{15}^S \left( E_x E_z^* + E_z E_x^* \right) + i \beta^A \left( E_x E_y^* - E_y E_x^* \right)$$

(1)

$$J_y = \beta_{22}^S \left( E_x E_y - E_y E_x \right) + \beta_{15}^S \left( E_y E_z^* + E_z E_y^* \right) + i \beta^A \left( E_y E_z^* - E_z E_y^* \right)$$

(2)

$$J_z = \beta_{31}^S \left( E_x E_z^* \right) + \beta_{31}^S \left( E_y E_y^* \right) + \beta_{33}^S \left( E_z E_z^* \right)$$

(3)

For a beam propagating along $z$ with polarization amplitude components $E_{x(y)} = A_{x(y)} e^{-i(kz + \varphi_{x(y)})}$ we have:

$$J_x(z) = I \beta_{22}^S \cos \Delta \varphi(z)$$

(4)

$$J_y = \beta_{22}^S \left( A_x^2 - A_y^2 \right)$$

(5)

$$J_z = \beta_{31}^S I$$

(6)

with $I = A_x^2 + A_y^2$ the total power density and $\Delta \varphi(z) = \varphi_y(z) - \varphi_x(z)$ the phase difference between the two polarization components.
The first component, $J_x$, is zero for a circularly polarized beam like the one here used in the Tardy setup because in this case $\Delta \varphi = \pi/2 = \varphi_0$. This result is in agreement with Neumann’s principle, since a circular polarization cannot break the three-fold symmetry of the crystal inducing a current along one specific direction other than the $z$ axis. However, as the beam propagates along the sample, the circular polarization prepared at the input of the sample (see Fig. 1) is in general transformed into elliptical along the sample, either due to the birefringence $\Delta n = n_x - n_y = n_3^x r_{22} E_2$ induced by the external electric field applied along the $y$-axis, either due to other disturbances or misalignment of the propagation direction of the beam with respect to the $z$ axis. In this case the phase difference increases with the propagation distance $z$:

$$\Delta \varphi(z) = \varphi_0 + z \frac{2\pi}{\lambda} \Delta n$$

In the worst case, for $z \frac{2\pi}{\lambda} \Delta n = \pm \pi/2$, the phase difference becomes equal to 0 or $\pm \pi$. The beam polarization would then be linear and $|J_x|$ would be maximal. The maximum attainable internal field generated at equilibrium by such a current is given by:

$$E_{\text{max}}^{\text{PG}} = I \beta_{22}/\sigma$$

where $\sigma$ is the photoconductivity. Since $\sigma$ is here taken to be proportional to $I$, $E_{\text{max}}^{\text{PG}}$ is not dependent on the beam intensity. It should be noted here that this is a rough majorant because the externally applied field, after being switched on, decreases monotonously to zero.

Figure 1: Polarization state of the beam at the input and output faces of the sample. The PG current increases along the sample as the polarization evolves.
Figure 2: (a) optical indicatrix in the $Oxy$ plane of a LN sample with an electrical field along $y$. (b) Same with an electrical field along $x$. (c) Polarization ellipse after the sample considering an electro-optic change of the indicatrix as depicted in (a). (d) Same, considering a change of the indicatrix as in (b).

due to the screening effects mentioned in the paper. Thus the polarization has the tendency to go back to circular all along the sample and the PG effects are thus frustrated during the measurement.

On the other hand, the $J_y$ component remains always close to zero as long as the amplitude of the two polarization components remains close to each other (see eq. (5)), as it is the case here. Finally, the $J_z$ component of the PG current creates some charges on the input and output faces of the sample that are in air and separated of several millimeters. We can therefore safely consider that they are screened by ambient charges before they can buildup a significant field and in the following we will neglect this contribution.

In summary we have to evaluate the potential effect of an $x$-directed $E_{PG}$ space charge with an upper limit $\ll E_{max}^{PG}$ compared to the externally applied field. Unfortunately, to the best of our knowledge, there are not detailed data on the value of the $\beta_{22}^{S}$ PG coefficient in undoped or Zn-doped LN, so we cannot perform an $a$ priori estimate. However we empirically determine that the PG-generated field is not perturbing our measurement as it
follows.

Taken into account the form of the electro-optic tensor of lithium niobate it can be seen that, while the externally applied field $E_x$ squeezes the optical indicatrix along the $\hat{x}$ and $\hat{y}$ directions, the internal field directed along $\hat{x}$ does the same but along the diagonal directions $\hat{u} = \hat{x} + \hat{y}$ and $\hat{v} = \hat{x} - \hat{y}$ (see Fig. 2 (a) and (b)). In our setup the polarization reading is achieved by means of a quarter wave plate placed after the sample and oriented at 45 degrees, thus along the diagonals $\hat{u}$ or $\hat{v}$. This design guarantees the maximum sensitivity to birefringence changes of the type described in Fig. 2 (a). However, for an applied field along $x$, the situation is as depicted in Fig. 2 (b). In those conditions the quarter wave plate is no longer working correctly and we should observe a marked change in the response function of our setup corresponding to a measurable alteration of the extinction ratio $I_{max}/I_{min}$ after the analyzer.

We checked several times the extinction ratio of our system, in different illumination conditions and at different times, with and without an applied field on both the samples: in all cases we did not notice any significant variation in the contrast, which proves that the field along $x$ is negligibly small. Thus we conclude that our experiment is sensitive only to the externally applied field.