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Interferometric measurement of the pyroelectric coefficient in lithium niobate

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We report the measurement of pyroelectric field in LiNbO₃ by introducing a new method, based on optical interferometry. The pyroelectric coefficient of the congruent LiNbO₃ crystals has been determined by this method and the value obtained for it, $\sim 95 \mu\text{C}/(\text{m}^2 \cdot \text{K})$, is consistent with previous results reported in literature, obtained by other methods. © 2013 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4788696>]

I. INTRODUCTION

The pyroelectric effect consists in a temporary voltage generation by changing the temperature of a pyroelectric material. This is obtained due to a change in the spontaneous polarization which has a material dependent sensitivity. The sensitivity of the effect can be characterized by the pyroelectric coefficient (p) defined as the spontaneous polarization (P_s) change due to temperature (T) change

$$p = \frac{\partial P_s}{\partial T}. \quad (1)$$

The pyroelectric effect is important for many applications like sensing, infrared imaging,¹ X-ray generation by electron acceleration,² or particle trapping.³ In this paper, we focus on the pyroelectric properties of electro-optic crystals like lithium niobate (LiNbO₃, or LN for short). LN crystals are already widely used in photonics applications, for electro-optic modulation and parametric frequency generation. Recently, the pyroelectric effect in LN was used to demonstrate applications like pyrolitons for light guiding^{4,5} or the photocatalytic disinfection.⁶

Different methods, electrical⁷⁻¹¹ or optical,¹² were previously used to measure the pyroelectric coefficient of LN. The results are spread in the range (-104 to -40) $\mu\text{C}/(\text{m}^2 \cdot \text{K})$. With a very small temperature change of only 1 K, an electric field of the order of kV/cm can be generated. It is important to accurately know the value of the pyroelectric coefficient since high fields generated by temperature change can significantly enhance light-Induce refractive index changes^{13,14} or can lead to undesired effects (electric discharges between crystal faces) that can affect the optical quality of the samples.

When using electrical methods to measure the pyroelectric coefficient, very good electrodes on the crystal surfaces are required. The electrodes can also be an impediment for the use of the investigated crystals in other experiments.

We introduce a new, simple, and fast all-optical method, based on interferometry, to measure the pyroelectric coefficient. By applying this method to a congruent LN crystal, we

measure the pyroelectric field generated inside the crystal for small temperature changes (units of K) and we determine its pyroelectric coefficient. We take into account the thermo-optic and the thermal expansion effects and minimize the photorefractive-photovoltaic effect that can affect the measurement of the pyroelectric coefficient.

This method can also be applied to the investigation of pyroelectric effect in other electro-optic materials.

II. THE EXPERIMENTAL SETUP AND METHOD

The experimental setup for the measurement of pyroelectric field and determination of the pyroelectric coefficient is shown in Fig. 1. It consists of a Mach-Zehnder interferometer with the crystal introduced in the signal arm of the interferometer.

A He-Ne laser at 632.8 nm wavelength was used as light source in the interferometer. Two polarizers (P1 and P2) were used to control the power and the polarization of the beams. A temperature controller coupled to a Peltier element (for heating) and to a thermistor (temperature sensor) is used to set the crystal temperature. The crystal is placed directly on the Peltier element and is covered with a thermal insulating plate on the top to minimize the temperature gradient along the c -axis of the crystal (vertical direction). The thermistor is fixed inside a small aluminum support, placed on the Peltier element, very close to one lateral side of the crystal. Beam splitters BS1 and BS2 consist of thick identical optical glass plates that ensure equal intensities of the interfering beams (signal and reference) and, consequently, a high visibility of the interference fringes. A CCD camera is used to acquire fringe images at a rate of 2 frames per second.

The shift of the interference fringes is determined by comparing each acquired frame with the previous frame. This shift is extracted in pixels from the acquired images and it is converted in radians by comparing it with the measured grating spacing that corresponds to a phase difference of 2π radians. The measured optical phase shift is used to determine the refractive index change. In LN crystals, this change is due to both the pyroelectric and thermo-optic effects. Due to the fact that LN crystal is smaller than the Peltier element, a small but not negligible contribution to the measured phase shift is given by the refractive index change of air near the crystal. This contribution is due to light passage through the

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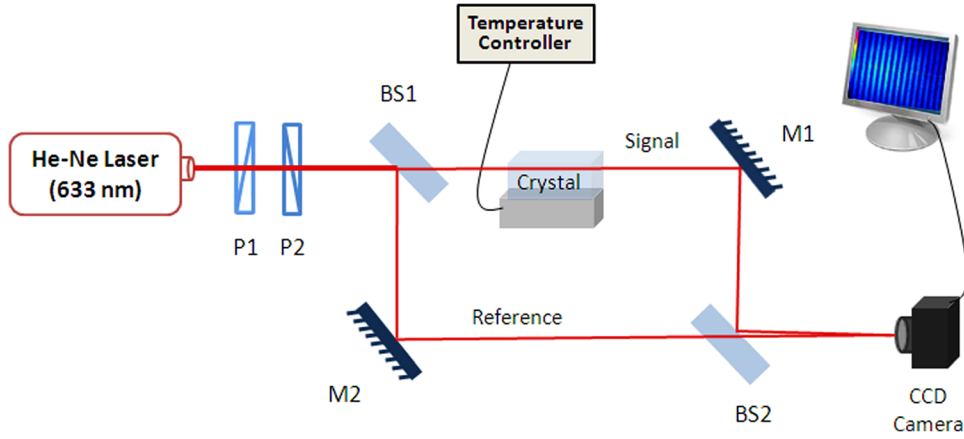


FIG. 1. The experimental setup.

heated air on the propagation direction, before the entrance face and after leaving the crystal. All the experiments were done in the same ambient conditions.

The optical phase change is given by

$$\Delta\Phi = \frac{2\pi}{\lambda} (\Delta n_{\text{LN}} \cdot L_{\text{LN}} + \Delta n_{\text{air}} \cdot L_{\text{air}}), \quad (2)$$

where λ is the wavelength of the interfering beams, Δn_{LN} is the refractive index change of the crystal, L_{LN} is the crystal length, Δn_{air} is the refractive index change of the air, and L_{air} is the length of the light path through the air region heated by the Peltier element. The refractive index change of the crystal is given by

$$\Delta n_{\text{LN}} = \Delta n_{\text{py}} + \Delta n_{\text{to}}, \quad (3)$$

where Δn_{to} is the thermo-optic contribution to the refractive index change and Δn_{py} is the pyroelectric contribution. Δn_{py} is given by

$$\Delta n_{\text{py}} = -\frac{1}{2} n^3 r_{\text{eff}} E_{\text{py}}, \quad (4)$$

where n is the bulk refractive index, r_{eff} is the electro-optic coefficient corresponding to the incident light polarization, and E_{py} is the pyroelectric field generated inside the crystal by the change of its temperature. In our experiments, we use extraordinary polarized (e-pol) light and the corresponding r_{eff} is r_{33} .

Δn_{to} is given by

$$\Delta n_{\text{to}} = \frac{\partial n_{\text{LN}}}{\partial T} \cdot \Delta T. \quad (5)$$

The thermo-optic contribution can be determined from temperature dependent Sellmeier equations.^{15,16} It was also measured in Ref. 17 and it is $\sim 4 \times 10^{-5} \text{ K}^{-1}$, at room temperature. The dependence of the thermo-optic coefficient on temperature, for e-pol light is¹⁷

$$\frac{\partial n_e}{\partial T} = (-2.6 + 22.4 \cdot 10^{-3} T) \cdot 10^{-5} \text{ K}^{-1}. \quad (6)$$

Similarly, the thermo-optic contribution of the heated air is given by

$$\Delta n_{\text{air}} = \frac{\partial n_{\text{air}}}{\partial T} \cdot \Delta T. \quad (7)$$

Using Eqs. (4), (6), and (7) in Eq. (2), one can obtain the pyroelectric field (E_{py}) generated inside the LN crystal

$$E_{\text{py}} = \frac{2}{n^3 r_{33}} \frac{\partial n_{\text{LN}}}{\partial T} \cdot \Delta T - \frac{1}{L_{\text{LN}} n^3 r_{33}} \left(\frac{\Delta\Phi \cdot \lambda}{\pi} - 2 \frac{\partial n_{\text{air}}}{\partial T} \cdot L_{\text{air}} \cdot \Delta T \right). \quad (8)$$

The pyroelectric tensor of LN has only one non-zero component that results in an electric field only in the direction of the crystal c -axis. Using this component of the pyroelectric coefficient, the pyroelectric field can be written as

$$E_{\text{py}} = -p \frac{\Delta T}{\epsilon_0 \epsilon_r}, \quad (9)$$

where the pyroelectric coefficient p is given by Eq. (1), ϵ_0 is the free space permittivity, and ϵ_r is the low frequency dielectric permittivity of the material. Thus, the pyroelectric coefficient can be determined from the slope of the linear dependence $E_{\text{py}} = f(\Delta T)$ (Eq. (9)).

III. EXPERIMENTS AND DISCUSSION

Several precautions have been taken to accurately measure the pyroelectric field and to determine the pyroelectric coefficient.

The wavelength of the light source in the interferometer was selected in order to have a low absorption¹⁸ and consequently to reduce photorefractive-photovoltaic effects. The interference fringes were oriented parallel or at very small angle with the c -axis of the LN crystal in order to neglect charge transport mechanisms responsible for the photorefractive-photovoltaic effects¹⁹ and to increase their stability. Furthermore, we work at very low power of the laser source, around 100 nW. The beam waist at FWHM of $\sim 3.6 \text{ mm}$ results an irradiance of $\sim 1 \mu\text{W}/\text{cm}^2$. At this level of irradiance, the parasitic photorefractive-photovoltaic effect is negligible for the time intervals of our experiments (1–2 min). This time interval is sufficient for the crystal to reach the thermal equilibrium. A short time interval for each

measurement is also beneficial to avoid the pyroelectric field decay.⁵

Another factor that could affect the measurement is the thermal expansion of the crystal. Thermal expansion on the propagation direction introduces a phase change due to the crystal length modification, but this factor is negligible in LN for small temperature changes.²⁰ Thermal expansion in the direction of the c -axis contributes to the secondary pyroelectric effect.²¹ The measured pyroelectric coefficient has a contribution from both primary and secondary pyroelectric effects.

The LN crystal sample is a piece ($4.83 \times 9.34 \times 2$ mm³) from a z -cut commercially available congruent LN wafer. The size of the Peltier element is 16×16 mm². Since the crystal is smaller than the Peltier element (crystal length is 4.83 mm on the propagation direction), the thermo-optic effect in air has an influence on the total fringe shift as discussed above. We measured the thermo-optic coefficient of air by heating the Peltier element and analyzing the fringe shift in the absence of the crystal. The measured value is -0.91×10^{-6} K⁻¹. This value is close to the values found in literature, -0.87×10^{-6} K⁻¹, for the temperature interval (293–313) K,²² and -1×10^{-6} K⁻¹, for the interval (273–293) K.²² In our case, the initial room temperature was ~ 294 K.

A typical temporal evolution of the phase variation, when heating the LN crystal with $\Delta T = 3$ K, is shown in Fig. 2.

The fringe recording is started several seconds before starting the heating ($t = 7$ s in Fig. 2), in order to estimate the noise introduced in the phase change by vibrations. The temperature is increased at a rate of ~ 0.1 K/s. The heating temperature reaches a maximum at $t \sim 38$ s that is higher than the set temperature change of $\Delta T = 3$ K, due to the typical temperature controller operation mode. The value considered for the fringe shift is after the temperature stabilization, when the crystal reaches the thermal equilibrium ($t > 65$ s in Fig. 2). Typical fringe patterns for the initial state ($t = 0$ s) and for the final steady-state ($t > 65$ s) are illustrated in Fig. 3.

Several experiments at different temperature changes were done, starting the heating from the same initial temperature of the crystal. The temperature change was in

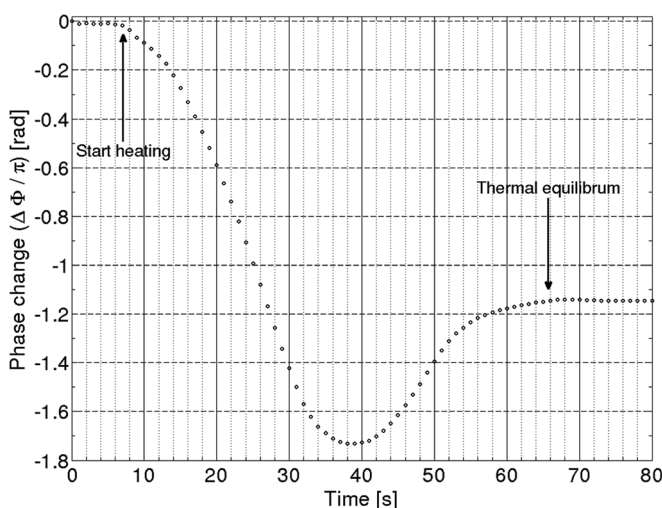


FIG. 2. Phase change due to heating of the LN crystal with $\Delta T = 3$ K.

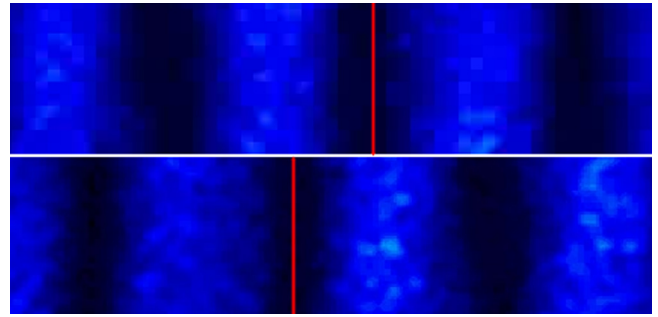


FIG. 3. Typical shift of fringe pattern between the initial state and the final steady-state, for $\Delta T = 3$ K.

the range of 1° to 10° . Higher heating temperatures may result in electric breakdowns on the crystal lateral faces due to the very high electric field generated. For example, by heating the crystal with only 10° , an electric field of the order of 40 kV/cm can be obtained. The corresponding pyroelectric field, computed at each ΔT using Eq. (8) and the measured value of the phase shift, is shown in Fig. 4. As stated above, we can determine the pyroelectric coefficient p by fitting the temperature dependence of the pyroelectric field and using Eq. (9).

To compute the pyroelectric field and to determine the pyroelectric coefficient, we need the values of the electro-optic coefficient, the refractive index, and the dielectric permittivity in Eqs. (8) and (9). We considered the value of 32.2 pm/V (Refs. 23 and 24) for r_{33} and a value of 0.016 (pm/V) \cdot K⁻¹ (Ref. 25) for its temperature variation. The refractive index was computed using Sellmeyer equations from Ref. 15 and the dielectric permittivity was taken as 28.7.²⁶

Considering the error sources (heating temperature, length of the heated air along the propagation direction of the signal beam, pixel error in phase shift), the total error in the measurement of the pyroelectric field is computed and is shown as error bars in Fig. 4.

By fitting the temperature dependence of the pyroelectric field with a linear function (Fig. 4), we obtained the

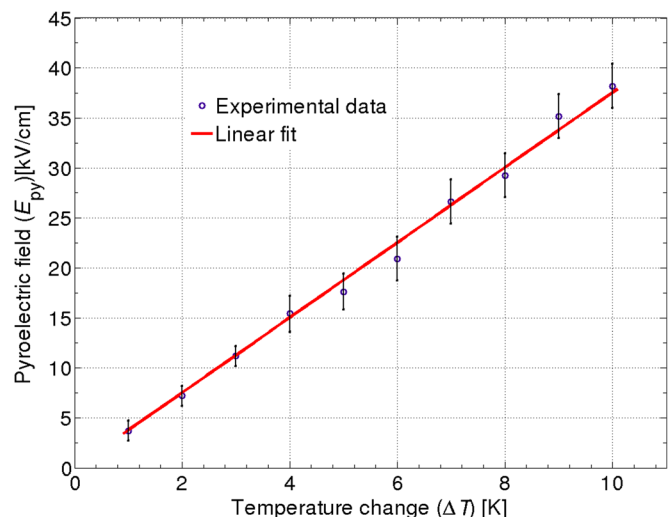


FIG. 4. Pyroelectric field dependence on the temperature change.

value of the LN pyroelectric coefficient, $p = (-95.4 \pm 5.8) \mu\text{C}/(\text{m}^2 \cdot \text{K})$. This value is consistent with the recently obtained result of $-103.9 \mu\text{C}/(\text{m}^2 \cdot \text{K})$ measured in congruent LN by other optical method.¹²

IV. CONCLUSIONS

We have measured the pyroelectric field and determined the pyroelectric coefficient of congruent LN crystals, by using a new, simple, and fast all-optical method based on Mach-Zehnder interferometry. The value of the pyroelectric coefficient is consistent with the results previously reported in literature, obtained by other methods. The optical method introduced in this paper can be also applied to the investigation of the pyroelectric effect in other electro-optic materials.

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