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Experimental investigation of the anisotropy of quadratic electrooptic effect in ADP

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Measurements of the quadratic electrooptic coefficient g_{1212} of ammonium dihydrogen phosphate (ADP) were made. The coefficient $g^* = g_{1111} - g_{2211} - 2g_{1212}$ related to the anisotropy of electrooptic effect is determined. The value obtained for the coefficient is $g^* = -15.8\times10^{-20}$ m^{2}V^{-2}. This shows that the anisotropy of quadratic electrooptic effect in ADP is stronger than that estimated previously for potassium dihydrogen phosphate.

Keywords: quadratic electrooptic effect, $NH_4H_2PO_4$.

1. Introduction

It is known that in mechanically isotropic materials, the elastic moduli c_{ijkl} fullfil the condition $c_{1111} - c_{2211} - 2c_{1212} = 0$ [1,2]. Analogously, in electrooptically isotropic media the relationship between components of the quadratic electrooptic (Kerr effect) tensor g_{ijkl} is given by $g_{1111} - g_{2211} - 2g_{1212} = 0$ [3]. Thus, the coefficient $g^* = g_{1111} - g_{2211} - 2g_{1212}$ may be useful to describe the electrooptical anisotropy of a material. The coefficient g^* has been previously employed in the investigation of the nature of nonlinearities responsible for electrooptic properties. For example, it has been considered in analyses of relationships between the linear electrooptic effect and the quadratic one in oxygen octahedra perovskites [4,5]. Moreover, an attempt has been made to use g^* to explain the behaviour of refractive indices and electrooptical coefficients of $LiNbO_3$ and $LiTaO_3$ in the ferroelectric phase [6]. These properties have been discussed in terms of nonlinear responses of the NbO_6 and LiO_6 (TaO_6 and LiO_6) octahedra.

The aim of this work is to determine the value of $g^* = g_{1111} - g_{2211} - 2g_{1212}$ in the $NH_4H_2PO_4$ (ADP) crystal.

2. Method

The quadratic electrooptic coefficients g_{1111} and g_{2211} of ADP have been previously determined [7,8]. The values of the linear coefficients are listed in Ref. 9. However, the value of g_{1212} is not known yet. This coefficient may be measured in two different configurations, namely parallel and perpendicularly to the optic axis, for example: $\sigma = (0,0,1)$ and $\mathbf{E} = (E,E,0)/\sqrt{2}$ or $\sigma = (1,\bar{1},0)/\sqrt{2}$ and $\mathbf{E} = (E,E,0)/\sqrt{2}$, where the intended light beam direction is described by the unit vector $\boldsymbol{\sigma}$ and $\mathbf{E}$ is the modulating electric field. When the first configuration is satisfied exactly, the ideal value of the modulation index $A_{2\Omega}^{id}$ due to the quadratic electrooptic response derived on the assumption of no inaccuracies is related to g_{1212} by

$$\left|g_{1212}\right| = \frac{\lambda A_{2\Omega}^{id}}{\pi L E_0^2 n_o^3 (1 + A_{2\Omega}^{id})}, \tag{1}$$

where λ is the wavelength, L is the crystal length, E_0 is the amplitude of the modulating electric field, and n_o is the ordinary refractive index. The second configuration allows the absolute value of the effective coefficient g_{ef} consisting g_{1212} to be measured

$$\left|g_{ef}\right| = \frac{2\lambda A_{2\Omega}^{id}}{\pi L E_0^2 (1 + A_{2\Omega}^{id})}, \tag{2}$$

where

$$\begin{aligned} g_{ef} &= n_e^3 g_{3311} - \frac{1}{2} n_o^3 (g_{1111} + g_{2211}) \\ &\quad - n_o^3 g_{1212} + \frac{n_o^3 + n_e^3}{n_o^2 - n_e^2} n_o^2 n_e^2 r_{231}^2, \end{aligned} \tag{3}$$

r_{231} is the linear electrooptic coefficient and n_e is the extraordinary refractive index.

In real measurements, some inaccuracies in crystal cutting and alignment may appear. Considering errors that can result from experimental inaccuracies, it has been indicated recently that when a quadratic electrooptic coefficient of a uniaxial crystal may be determined either parallel or perpendicularly to the optic axis, the second configuration should be used [10]. This is of special importance in measurements of coefficients like g_{1212}, which are associated with rotations of the principal axes of the optical

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permittivity tensor. Furthermore, the calculations show that in these measurements an application of a dynamic modulating field significantly decreases the experimental error.

To compare, in the two possible configurations, the sensitivity of measurements for an imprecise crystal cutting and alignment, we employed the Jones calculus extended recently for a modulated double refracted light beam propagating in an electrooptic uniaxial crystal [11]. In our calculations, the crystal was considered to be cut in the form of a right parallelepiped with intended orientation of its faces parallel to the *x, y*, and *z* axes. The intended direction of the incident light with $\lambda = 0.63$ nm was taken to agree with the +*z* axis. In the analysis, the distribution of the light amplitude in the cross-section of the beam was assumed to be Gaussian. The radius of the beam was taken 0.5 mm. The angles β_c and γ_c describing the inaccuracies in the crystal cutting specify the rotations of the *xyz* system about the +*x* and +*y* axes, respectively. Thus, the angles β_c and γ_c are defined by the transformation **b** from the intended crystal cutting orientation system *xyz* to the *x'y'z'* axes related with the faces of imperfectly cut crystal. This transformation is given by [11]

$$\mathbf{b} = \begin{bmatrix} \cos\gamma_c & 0 & -\sin\gamma_c \\ 0 & 1 & 0 \\ \sin\gamma_c & 0 & \cos\gamma_c \end{bmatrix} \begin{bmatrix} 1 & 0 & 0 \\ 0 & \cos\beta_c & \sin\beta_c \\ 0 & -\sin\beta_c & \cos\beta_c \end{bmatrix}. \quad (4)$$

To discuss the influence of the inaccuracies on the results of measurements of quadratic electrooptic coefficients we employed the following ratio

$$\Delta_{2\Omega} = \left| A_{2\Omega} - A^{id}_{2\Omega} \right| \Big/ A^{id}_{2\Omega}, \quad (5)$$

where $A_{2\Omega}$ is the quadratic electrooptic response affected by the inaccuracies.

Examples of the results obtained allowing for inaccuracies in the crystal cutting and assuming that the light beam enters the crystal faces precisely perpendicularly are illustrated in Figs. 1 and 2.

The plots shown in Fig. 2 correspond to the case when the measurements are performed on the middle of the transmission characteristic of the system with the electrooptical crystal. When $\sigma = (0,\bar{1},0)/\sqrt{2}$ such condition may be easily fulfilled employing the experimental technique described in Ref. 12. For the direction $\sigma = (0,0,1)$ a thin quarter-wave plate was assumed. The comparison of the plots presented in Figs. 1 and 2 confirms that the sensitivity for the inaccuracies of measurements performed with the light sent perpendicularly to the optic axis is much lower than that performed along this direction. An analogous conclusion may be drawn when the inaccuracies in the crystal alignment are considered. Thus, our measurements have been performed for the He-Ne laser beam propagating in the direction $\sigma = (1,\bar{1},0)/\sqrt{2}$.

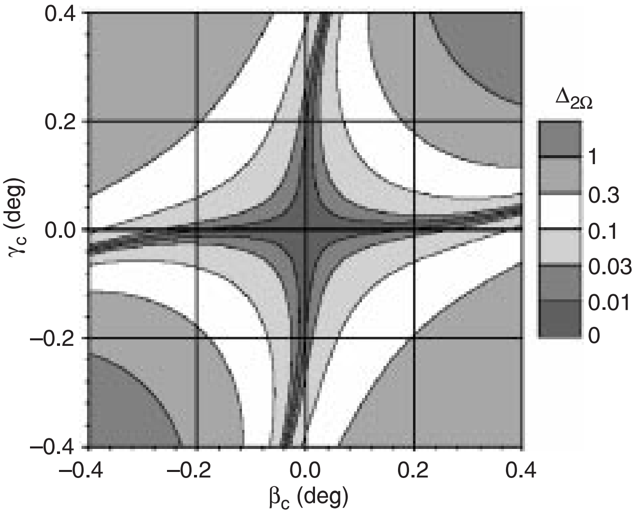

Fig. 1. Relative error $\Delta_{2\Omega}$, as defined by Eq. (5), in the measure of the $|g_{1212}|$ coefficient caused by inaccuracies in the crystal cutting in the absence of inaccuracies in crystal alignment for $\sigma = (0,0,1)$ and $\mathbf{E} = (E,E,0)/\sqrt{2}$. The amplitude of modulating field is 1.2×10^6 Vm^{-1} and the crystal length is 25 mm.

3. Experimental

In our measurements we employed the dynamic polarimetric method. This technique, as described in Ref. 12, is based on the analysis of the relative light intensity modulated by the applied sinusoidal electric field. The amplitude of the modulating field of frequency 417 Hz was 1.2×10^6 Vm^{-1}. Like previously [12], instead of using a compensator, we obtained the optical bias providing measurements on the most sensitive linear part of the transmission characteristic of the system by taking advantage of the temperature dependence of the natural birefringence in the

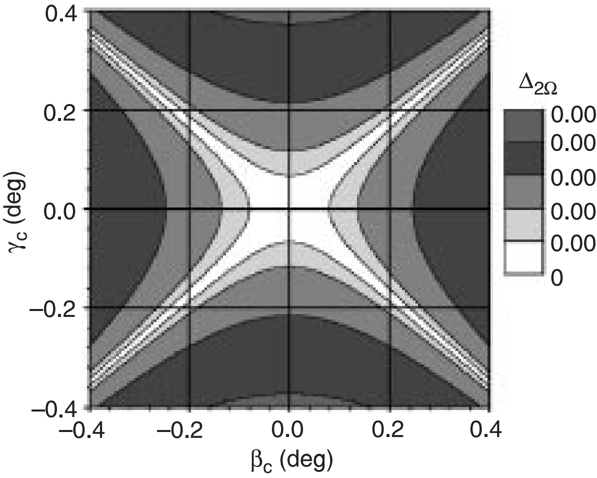

Fig. 2. Relative error $\Delta_{2\Omega}$, as defined by Eq. (5), in the measure of the $|g_{ef}|$ coefficient caused by inaccuracies in the crystal cutting in the absence of inaccuracies in crystal alignment for $\sigma = (1,\bar{1},0)/\sqrt{2}$ and $\mathbf{E} = (E,E,0)/\sqrt{2}$. The amplitude of modulating field is 1.2×10^6 Vm^{-1} and the crystal length is 25 mm.

measured crystal. The investigated ADP sample was cut in the form of parallelepiped of dimensions 25.1×25.1× 3.3 mm^3. The crystal was mechanically free.

The polarimetric technique allows only the absolute value of the electrooptic coefficient to be found. At temperature 295 K, our measurements gave for ADP $|g_{ef}| = (7.9 \pm 0.5)\times10^{-20}$ m^2V^{-2}.

4. Conclusions

The magnitudes and signs of the quadratic electrooptic coefficients g_{1111}, g_{2211} and g_{3311} appearing in Eq. (3) have been previously determined for ADP employing an actively stabilized Michelson interferometr [7]. Taking for g_{2211} and g_{3311} the correction factor from [8], the coefficients are as follows: $g_{1111} = -7.4\times10^{-20}$ m^2V^{-2}, $g_{2211} = -1.6\times10^{-20}$ m^2V^{-2} and $g_{3311} = -1.3\times10^{-20}$ m^2V^{-2}. The recommended value of r_{231} in ADP is -23.4×10^{-12} mV^{-1} [9]. These values, along with the measured $|g_{ef}|$ gave $g_{1212} = 5.0\times10^{-20}$ m^2V^{-2} or $g_{1212} = 9.5\times10^{-20}$ m^2V^{-2}, for the positive and negative signs of g_{ef}, respectively. To consider the sign of g_{ef} we took into account the bond polarizability model [13]. The nonlinear optical properties of KDP-type crystals are usually related to the response of the PO_4 tetrahedra. The bond polarizability approach applied to the PO_4 groups suggests that the coefficient g_{ef} is positive. This allows to obtain $g_{1212} = (5.0 \pm 1.1)\times10^{-20}$ m^2V^{-2} and $g^* = (-15.8 \pm 7.4)\times10^{-20}$ m^2V^{-2}.

Our measurements indicate that the anisotropy of the quadratic electrooptic effect of ADP is stronger than that in KDP for which $|g^*|$ may be estimated to be smaller than 6.0×10^{-20} m^2V^{-2} [7]. We hope that in future we will be able to relate the coefficient g^* to the crystal structure and explain if the fourth-order electrooptic effect contributes to the anisotropy of the quadratic electrooptic effect.

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