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ESTIMATIONS OF THE MAGNITUDE OF THE FOURTH-ORDER ELECTROOPTIC EFFECT IN KDP-TYPE CRYSTALS

An attempt to measure the fourth-order electrooptic effect in KH_2PO_4 *(KDP) and* $NH_4H_2PO_4$ *(ADP) is presented. Contrary to previously reported results of the order of magnitude* 10^{-30} m^4V^{-4}*, the fourth-order coefficient* $|g_{111111} - g_{221111}|$ *is found to be lower than* $5 \cdot 10^{-33}$ m^4V^{-4} *and* $1 \cdot 10^{-34}$ m^4V^{-4}*, respectively. In addition, the effect of imperfect crystal cutting and alignment is investigated by means of calculations based on Jones calculus. It is found that even small inaccuracies may lead to an apparent fourth-order electrooptic effect comparable in magnitude to the previously reported values.*

Keywords: fourth-order electrooptic effect, KH_2PO_4, $NH_4H_2PO_4$.

1. INTRODUCTION

Results of measurements of the fourth-order electrooptic coefficients have been previously reported for some crystals including $BaTiO_3$ [1] and members of the KDP (KH_2PO_4) family (see, e.g. [2-4]). The first published results for the fourth-order effect in KDP type crystals have been obtained employing static fields and indicate very large values of the order of magnitude 10^{-30} m^4V^{-4}. However, these results have not been confirmed by further attempts to measure the effect by dynamic means [5,6]. One notes that somewhat similar situation may be observed for the quadratic electrooptic effect, where apparent coefficients obtained by static methods and due to an imperfectly cut or aligned crystal are sometimes three orders of magnitude larger than results of dynamic measurements [7,8]. The theoretical results presented recently for the quadratic electrooptic effect have shown that the sensitivity of measurements performed by static means for the inaccuracies is much higher than in methods employing a sinusoidal electric modulating field. Corresponding nonlinearities may lead to apparent nonlinear electrooptic effects [9-11].

The fourth-order effect is important from the point of view of the relationship between the spontaneous birefringence and spontaneous antipolarization in the low-temperature phase of $NH_4H_2PO_4$ (ADP) [12]. One notes that a significant contribution of the fourth-order electrooptic effect would affect this relationship. Thus, nonlinear electrooptic properties of ferroelectric and antiferroelectric crystals are of interest as related to the nature of the spontaneous birefringence in their low-temperature phases. Moreover, the fourth-order electrooptic effect is important in analyses of the temperature dependencies of intrinsic (*i.e.* defined in terms of polarization) quadratic electrooptic coefficients in ferroelectrics and ferroelectrics [16]. Nonlinear electrooptic effects may also lead to nonlinear responses of various technical devices employing electrooptic crystals, therefore, are important in applications.

The aim of this work is to investigate the magnitude of the fourth-order electrooptic coefficient $g_{111111} - g_{221111}$ in KDP and ADP crystals.

2. EXPERIMENTAL

We have employed the dynamic polarimetric technique described previously (see, e.g. references [12,13]) and based on the harmonic analysis of light modulated by the sinusoidal modulating voltage $U(t) = U_0 \sin(\omega t)$ using the electric field of frequency 417 Hz. Such a frequency is much lower that any piezoresonance frequency in our samples, so that the crystals resting freely on their supports could be regarded as mechanically free. The samples under investigation were positioned so that the He-Ne laser beam propagated along their optical axes. Measurements were made for several such light paths across the entrance face of each crystal. The modulator used in measurements was biased to the middle of its transmission characteristic by a quarter-wave plate. To apply the electric field parallel to the [100] direction, the faces of samples were coated with conducting silver paint. The samples were placed in a bath containing a silicon oil having checked previously that the contribution of its Kerr effect due to the fringing field is negligible. The measurements were performed at temperature 295 K. The use of the lock-in amplifiers enabled sensitive measurements of the response of the system to be made on frequencies ω, 2ω and 4ω. The readings on the fundamental frequency ω were useful to provide precise orientations of the investigated crystals. Our measurements were made on crystals with dimensions 50 × 50 × 5 mm^3.

For the configuration under consideration and in the absence of any errors in crystal cutting or alignment the electric field induced birefringence in crystals belonging to the point symmetry $\bar{4}2m$ group is given as [14]

$$\Delta n(t) \approx \frac{n_\mathrm{o}^3}{2}\left[(g_{1111} - g_{2211})E(t)^2 + (g_{111111} - g_{221111})E(t)^4\right], \tag{1}$$

where n_o is the ordinary refractive index and g_{ijkl} and g_{ijklmn} are components of the quadratic and fourth-order electrooptic tensors, respectively. Thus, the absolute value of the fourth-order coefficient $g_{111111} - g_{221111}$ is related to the response $U(4\omega)$ of the modulator used in measurements on the fourth harmonic by

$$\left|g_{111111} - g_{221111}\right| = \frac{8\lambda d^4 U(4\omega)}{\pi n_\mathrm{o}^3 l U_\mathrm{c} U_0^4}, \tag{2}$$

where d is the crystal thickness, l its length and U_c is the voltage measured the constant component of the light intensity.

With sensitivity of our apparatus allowing to detect the fourth-order electrooptic coefficient $|g_{111111} - g_{221111}|$ of magnitude $2\cdot 10^{-34}$ m^{4}V^{-4} and $1\cdot 10^{-34}$ m^{4}V^{-4}, for KDP and ADP, respectively, we have not been able to observe any response. Thus, this effect should be lower.

3. THE APPARENT FOURTH-ORDER ELECTROOPTIC EFFECT

In real measurements some inaccuracies in crystal cutting and alignment may appear. To explain the discrepancy between our results and the former obtained by static means, we have performed computer simulations of measurement conditions of the fourth-order coefficients. In this we have followed the approach employed previously for the linear and quadratic electrooptic effects [8-11,15]. We have applied the Jones calculus for a modulated double-refracted light beam propagating in an electrooptic uniaxial crystal [11] employing the angles describing the inaccuracies as shown in Fig. 1. Examples of our results obtained for the fourth-order apparent electrooptic effect are plotted in Fig. 2.

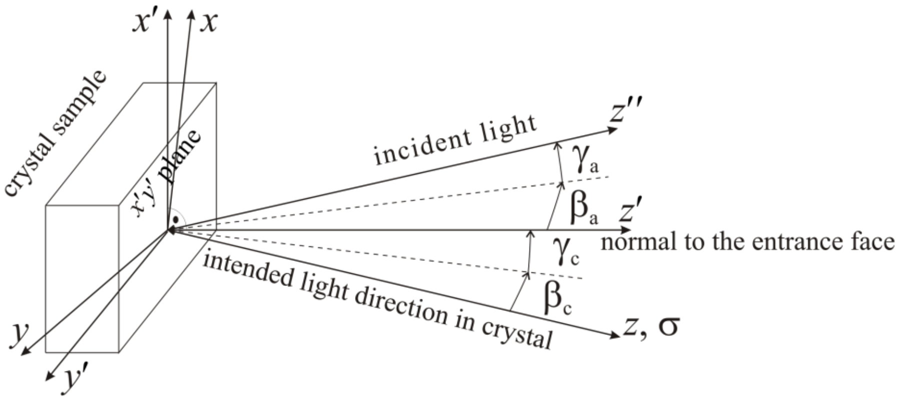

Fig. 1. Definition of the angles β_c and γ_c describing the inaccuracies in the crystal cutting and β_a and γ_a related to its inaccurate alignment

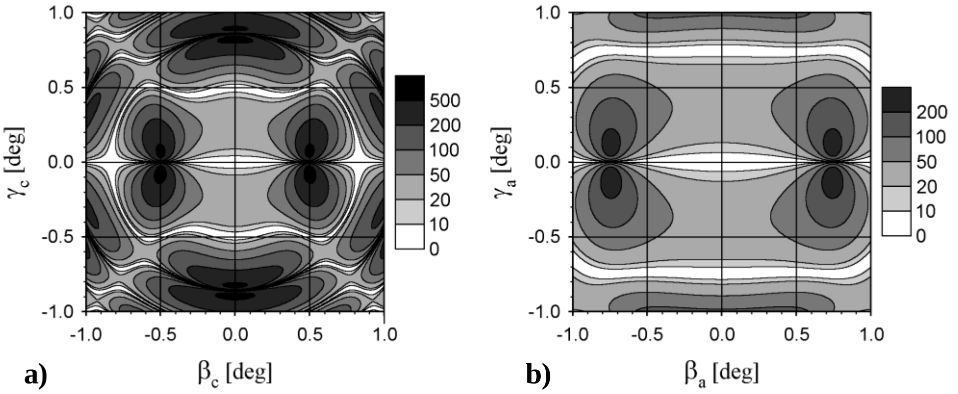

Fig. 2. Calculated values of the apparent fourth-order coefficient $|g_{111111} - g_{221111}|$ in 10^{-33} m^4V^{-4} in KDP obtained by assuming the use of the static electric field of strength up to 10^6 Vm^{-1}. The radius of the light beam of the uniform intensity is 0.5 mm, the wavelength λ = 0.63 μm and the crystal length is l = 5 cm. The intended directions of the light beam and the electric field are $\boldsymbol{\sigma} = (1,0,0)$ and $\mathbf{E} = (E,0,0)$. Considered examples: a) inaccuracies in the crystal cutting in the absence of inaccuracies in the crystal alignment, b) inaccuracies in the crystal alignment in the absence of inaccuracies in the crystal cutting

4. DISCUSSION

In our opinion, earlier results that are not confirmed by our measurements stem from the lower-orders electrooptic effects in the presence of inaccuracies in the alignment or cutting of investigated samples. The plots presented in Fig. 2 suggest that analogously to some earlier measurements of the quadratic electrooptic effect, the previously determined very large fourth-order coefficients are apparent not the real ones.

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PRÓBA WYZNACZENIA WIELKOŚCI ELEKTROOPTYCZNEGO EFEKTU CZWARTEGO RZĘDU W KRYSZTAŁACH GRUPY KDP

Streszczenie

Wykorzystując dynamiczną metodę polaryzacyjno-optyczną przeprowadzono próbę pomiaru elektrooptycznego efektu czwartego rzędu w kryształach KH_2PO_4 (KDP) oraz $NH_4H_2PO_4$ (ADP). W przeciwieństwie do zmierzonych wcześniej przy wykorzystaniu metody statycznej i prezentowanych w literaturze wielkości współczynnika $|g_{111111} - g_{221111}|$ rzędu 10^{-30} m^{4}V^{-4}, otrzymane w pracy wyniki pozwalają stwierdzić, że współczynnik ten, odpowiednio dla kryształów KDP i ADP, jest mniejszy niż $5\cdot10^{-33}$ m^{4}V^{-4} oraz $1\cdot10^{-34}$ m^{4}V^{-4}. Analizując przyczyny rozbieżności otrzymanych wyników i tych otrzymanych wcześniej, rozważono wpływ błędów w wycięciu i orientacji wykorzystanych próbek krystalicznych na wynik pomiarów. Wykorzystując rachunek Jonesa pokazano, że takie niedokładności mogą prowadzić do znaczących nieliniowości w odpowiedzi układu pomiarowego, sugerując istnienie pozornego efektu czwartego rzędu, który może być nawet o kilka rzędów wielkości większy niż efekt prawdziwy.