Optica Applicata, Vol. XXXIII, No. 1, 2003

Measurement conditions of the quadratic electrooptic coefficients along the optic axis in uniaxial crystals

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Following our previous discussions of measurement accuracy of electrooptic coefficients in uniaxial crystals the light wave propagation along the optic axis of ammonium dihydrogen phosphate (ADP) in a field that bisects the X and Y crystallographic axes is considered. We analyse the effect of imperfection in crystal cutting and alignment by means of computer analysis based on the Jones calculus. It is confirmed that, for relatively small inaccuracies, the values of the quadratic electrooptic coefficients can be significantly larger when measured with a static field than those measured with a sinusoidal one.

Keywords: quadratic electrooptic effect, ADP crystal, Jones calculus.

1. Introduction

The electrooptic effect manifests itself as a change in the refractive index of a medium to which a static or low-frequency field is applied. The electrooptic coefficients are traditionally defined in terms of changes in the impermeability tensor B_{ij} induced in a crystal by an electric field $\mathbf{E}$

$$\Delta B_{ij} = r_{ijk}E_k + g_{ijkl}E_kE_l + \ldots \tag{1}$$

where r_{ijk} and g_{ijkl} are the linear and the quadratic electrooptic coefficients, respectively. The electrooptic effect finds extensive use in a variety of technological devices. Studies of electrooptic properties of crystals are also of interest from the point of view of understanding the nature of nonlinear susceptibilities that are related to the interaction of low-frequency electric fields with the crystal lattice [1]. However, the magnitudes of quadratic electrooptic coefficients of some crystals, especially of those which lack the center of symmetry, are still not well established. For example, despite the fact that the potassium dihydrogen phosphate (KDP) type crystals have received considerable attention because of their marked nonlinear properties, values of the quadratic electrooptic coefficients reported for members of this family of crystals differ by two or three orders of magnitude (see, *e.g.*, results listed in [1], [2]).

Previously, we have compared the accuracy of measurements of some linear and quadratic electrooptic coefficients performed employing static and dynamic fields along the optic axis of uniaxial crystals [3], [4]. It has been found that a greater potential for experimental errors exists in the measurement of those electrooptic coefficients that are associated with rotations of the principal axes of the optical permittivity.

The aim of this work is to analyse the accuracy of measurements of the quadratic electrooptic coefficient g_{xyxy} along the optic axis of ADP with static and dynamic electric field. We found that the use of a dynamic field greatly reduced the error.

2. Method

Our numerical calculations are based on the Jones calculus [5]. The light entering the crystal and that emerging from the modulator is described by one-column Jones vectors

$$\boldsymbol{\mathcal{E}}_0 = \begin{bmatrix} \mathcal{E}_{0x} \\ \mathcal{E}_{0y} \end{bmatrix}, \quad \boldsymbol{\mathcal{E}} = \begin{bmatrix} \mathcal{E}_x \\ \mathcal{E}_y \end{bmatrix}. \tag{2}$$

In the Jones approach, the polarization and the intensity of the light can be described by vectors which are composed of the x and y components of the optical frequency electric field vector. In terms of the Jones vector components the light intensity I is given by

$$I = |\mathcal{E}_x|^2 + |\mathcal{E}_y|^2. \tag{3}$$

According to the calculus, each optical element is described by a two-by-two matrix. The most general form of the Jones matrix is derived in [6]. The light emerging from the n-th plate in the series of n plane-parallel optical elements can be expressed as

$$\boldsymbol{\mathcal{E}} = \mathbf{J}_n \mathbf{J}_{n-1} \cdots \mathbf{J}_1 \boldsymbol{\mathcal{E}}_0. \tag{4}$$

Following experimental procedures employed previously to study the quadratic electrooptic effect in KDP-type crystals [4], the experimental set-up considered in our analysis consists of a polariser, a quarter-wave plate, a sample of ADP crystal, and an analyser. We assumed the electrodes to be deposited on the planes of the sample. Therefore, imperfections in the crystal cutting affect also the direction of electric field relative to the crystallographic axes

$$\begin{bmatrix} \mathcal{E}_x \\ \mathcal{E}_y \end{bmatrix} = \begin{bmatrix} 0 & 0 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} \cos^2\alpha_f + \sin^2\alpha_f e^{-i\Gamma} & \sin\alpha_f \cos\alpha_f (1 - e^{-i\Gamma}) \\ \sin\alpha_f \cos\alpha_f (1 - e^{-i\Gamma}) & \sin^2\alpha_f + \cos^2\alpha_f e^{-i\Gamma} \end{bmatrix} \frac{1}{2} \begin{bmatrix} 1 - i & 1 + i \\ 1 + i & 1 - i \end{bmatrix} \begin{bmatrix} \mathcal{E}_{0x} \\ 0 \end{bmatrix} \tag{5}$$

where α_f is the azimuth of a fast wave, and

$$\Gamma = \frac{2\pi l (n_s - n_f)}{\lambda} \tag{6}$$

is the phase difference between the slow and fast waves. In Eq. (6), n_f and n_s denote the refractive indices of the fast and slows waves, respectively, and l is the geometrical path of a light wave in the crystal. In our calculations, the numerical values of n_f and n_s and the azimuth of a fast wave α_f for a given direction of the light beam were found by employing the optical indicatrix. The crystal length was taken to be 1 cm and the light wavelength $\lambda = 0.63$ μm.

3. Effect of imperfection in crystal cutting and alignment

Assuming perfect alignment of a crystal of symmetry $\bar{4}2$m, for the configuration under consideration, where the light direction is (0,0,1) and $\mathbf{E} = (E, E, 0)/\sqrt{2}$, the electric field-induced birefringence is given by [2]

$$n_f - n_s = n_o^3\left(g_{xyxy} + \frac{1}{2}n_e^2 r_{yzx}^2\right)E^2 \tag{7}$$

where n_o and n_e are the ordinary and extraordinary refractive indices, respectively. Equation (7) shows that the electrooptic response, apart from the expected contribution due to the quadratic electrooptic coefficient, includes also a term in the square of the linear coefficient, even for a configuration for which symmetry vetoes a linear response. In ADP, where a large value of the linear electrooptic coefficient r_{yzx} is observed, this term becomes important. In our work we considered the effect of errors in cutting the crystal sample and also of the divergence of the light beam from the optic axis on measurement results. For these errors the theoretical expression (7) no longer applies.

We assumed the sample to be cut in the form of a right parallelepiped, with its axis, hereafter termed z' to avoid confusion, diverging from the optic axis z, as described by the angles β and γ in Fig. 1. Because of the deviation between z' and z axes, the faces of the parallelepiped, and therefore the electrodes on them, do not coincide with the crystallographic faces. Accordingly, the field in the parallelepiped has different components from these assumed in Eq. (7). In addition to the effect of angles β and γ

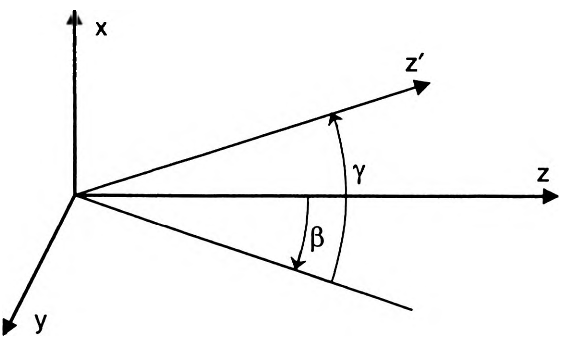
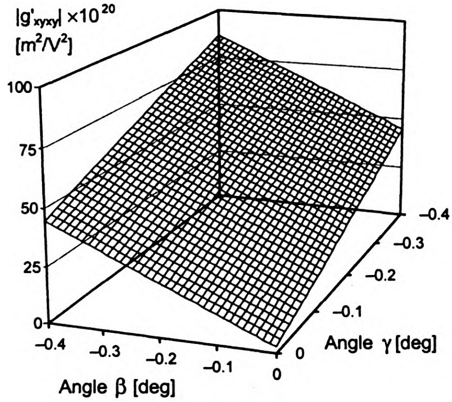


Fig. 1. Angles β and γ describing the inaccuracy in crystal cutting in terms of deviation of the axis z' of the crystal parallelepiped from its optic axis z.

on the emerging light intensity, a rotation about the z' axis was also allowed for. However, its effect was found to be negligible for the same range of angles used for β and γ. Two directions have been considered for the light beam: along the optic z axis and along the z' direction.

To illustrate the effect of inaccuracies in the crystal cutting, in Figs. 2 and 3 the dependences of the apparent quadratic electrooptic coefficient g'_{xyxy} on the angles β



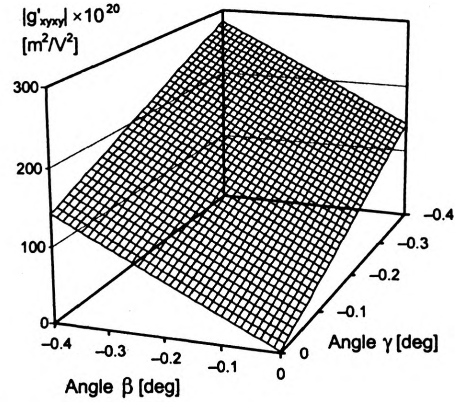


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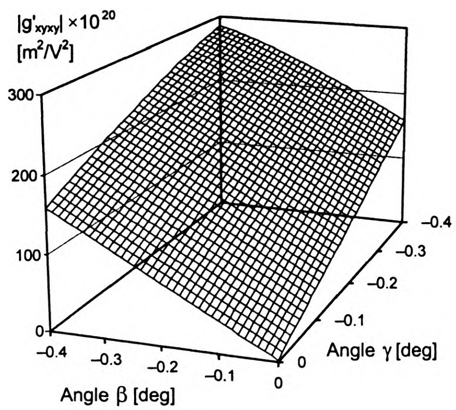
Fig. 2. Effect of inaccurate cutting of the ADP crystal on the apparent quadratic electrooptic coefficient g'_{xyxy} determined by the static method. The strength of the electric field is 3×10^4 V/m and the light wave propagates along the z direction.

Fig. 3. The same as in Fig. 2, but the strength of the electric field is 10^5 V/m.



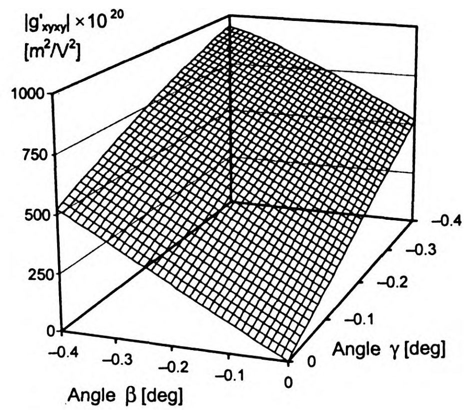


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Fig. 4. The same as in Fig. 2, but the strength of the electric field is 3×10^4 V/m and the light wave propagates along the z' direction.

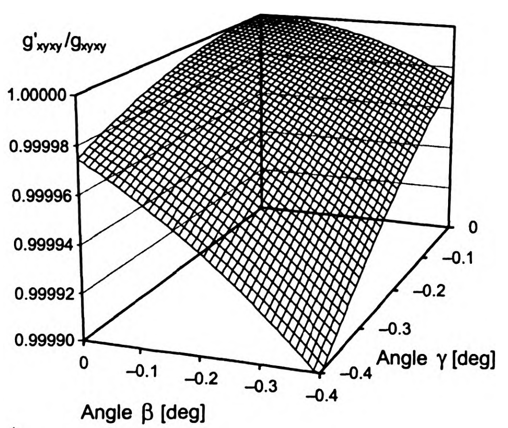
Fig. 5. The same as in Fig. 2, but the strength of the electric field is 10^5 V/m and the light wave propagates along the z' direction.

and γ are plotted. The coefficient g'_{xyxy} denotes the value that would be derived from the measured changes in the light intensity, namely

$$g'_{xyxy} = -\frac{\lambda A_{2\omega}}{\pi l n_o^3 E(1 + A_{2\omega})} \tag{8}$$

where $A_{2\omega}$ is the ratio of the modulator response on the second harmonic of modulating field to its d.c. component. Figures 2 and 3 show values of g'_{xyxy} for the static field of strength 3×10^4 V/m and 10^5 V/m, respectively. In our calculations we employed numerical values of the linear electrooptic coefficients of ADP from [7] and, when available, values of the quadratic coefficients from [3]. In case of those quadratic electrooptic tensor components that have been not measured yet, we assumed their numerical values to be $g_{xxzz} = g_{zzzz} = -2\times10^{-20}$ m^{2}V^{-2} and $g_{yzyz} = g_{xyxy} = (g_{xxxx} - g_{xxyy})/2 = -2.85\times10^{-20}$ m^{2}V^{-2}. However, either slight changes in their magnitudes or changes of their signs do not affect our conclusions.

The results obtained show that for inaccuracies in crystal cutting corresponding to angles $\beta = 0.4°$ and $\gamma = 0.4°$, the changes in the light intensity determined by static means lead to a value for g'_{xyxy} of nearly comparable magnitude as that measured in a static polarimetric experiment (see, *e.g.*, [4]). The comparison of the results plotted in Figs. 2 and 3 shows also that this apparent quadratic electrooptic coefficient obtained with static field depends strongly on the electric field strength. Considering the light propagation along the z' directions we found the results of the inaccuracies, as presented in Figs. 4 and 5, to be very significant as well.



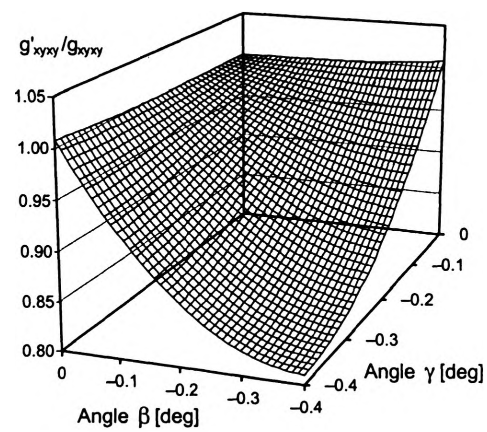


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Fig. 6. Effect of inaccurate cutting of the ADP crystal on g'_{xyxy}/g_{xyxy} for a sinusoidal field of amplitude 10^5 V/m when the light wave propagates along the z direction, g'_{xyxy} and g_{xyxy} are the apparent and true values of the quadratic electrooptic coefficients, respectively.

Fig. 7. Effect of inaccurate cutting of the ADP crystal on g'_{xyxy}/g_{xyxy} for a sinusoidal field of amplitude 10^5 V/m when the light wave propagates along the z' direction.

The errors in the g_{xyxy} coefficient presented in Figs. 2–5 can be dramatically reduced by employing a.c. fields. Figures 6–7 show for the same inaccuracies that the use of sinusoidal modulating field allows to obtain much smaller ratio of the apparent to true values of g_{xyxy}. The dependence of this ratio on the amplitude of a.c. field is very weak. Therefore, we present results for only one amplitude of the electric field.

4. Conclusions

The results obtained indicate that the error in the quadratic electrooptic coefficients responsible for the rotation of the principal axes of the optical permittivity around the z direction is relatively insensitive to the small deviation in the crystal cutting or alignment when a dynamic field is used. By contrast, the corresponding ratio g'_{xyxy}/g_{xyxy} for a static field is roughly two orders of magnitude. The computer calculations can explain incredibly large, *i.e.*, 6×10^{-17} m^2V^{-2}, value of the $|g_{xyxy}|$ coefficient determined previously for ADP by the static method.

Our calculations allow also to increase the accuracy of interferometric measurements of the electrostrictive coefficients in transmission [8].

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Received May 29, 2002