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UDC 539

EVALUATION OF NON-DIAGONAL COMPONENTS OF THE POCKELS TENSOR FOR PHOTOREFRACTIVE $Sn_2P_2S_6$ CRYSTAL

The technique of evaluation of the non-diagonal components r_{42} and r_{62} of the Pockels tensor for a monoclinic photorefractive crystal is developed and used for $Sn_2P_2S_6$.

K e y w o r d s: Pockels tensor, photorefractive crystal.

1. Introduction

Most part of photorefractive crystals have orthorhombic, trigonal, or higher symmetry, but several monoclinic photorefractives are also known. For example, tin hypothiodiphosphate ($Sn_2P_2S_6$, SPS) [1] and bismuth titanate [2] are monoclinic. The low symmetry of these crystals results in the necessity to use particular Cartesian frames to describe their dielectric, optical, and electro-optical properties that do not coincide with the standard crystallographic frame. This complicates the task of characterization for any new synthesized low-symmetry crystal.

At present, a special attention is given to SPS crystals (point group m). They have a large gain factor (up to 40 cm^{-1}) and a fast response (down to the 10-millisecond range) [3]. Being transparent in the red and the near-infrared, this material could be combined with powerful infrared laser sources and optical fibers. SPS crystals have been successfully used for the beam clean-up of high-power Nd^{3+} amplifier systems [4], the production of dynamic waveguides [5], the manipulation with a light pulse velocity [6], *etc.*

In spite of the fact that the electro-optical properties are of crucial importance for the photorefractive effect, the Pockels tensor of an SPS crystal has been studied incompletely.

All its nonvanishing components barring the non-diagonal components r_{42} and r_{62} were determined from the measurements of a crystal birefringence induced by an external electric field [7, 8].

Different photorefractive wave-mixing phenomena were also used to estimate all 10 component of the Pockels tensor of such a crystal [9–11]. The signs of components r_{42} and r_{62}, however, stay still unknown.

The purpose of this work is to determine both the values and the signs of r_{42} and r_{62} components of the Pockels tensor, which will complete our knowledge about the Pockels tensor. The technique is developed which allows extracting these data from the measurements of the gain factor dependence on the sample tilt angle. In Section 2, the appropriate recording geometries are discussed, and all crystal parameters that affect the gain factor are found. Then the implementation of the proposed technique for an SPS crystal is demonstrated. The experimental results are discussed and compared with calculations in Section 4.

2. Method

The techniques of evaluation of Pockels coefficients from the parameters of light self-difraction in a monoclinic photorefractive crystal [9, 11] are modified to estimate the non-diagonal components of tensor's second column.

The orientation of the recording beams in the crystal and their polarizations (what is called below "geometry") are selected in such a way that the beams interact via nonzero r_{42} or r_{62} components. The calculations allow expressing these unknown tensor components through the known components and the known experimental data.

As is known [12], the interference of two beams in a photorefractive crystal leads to the formation of a space charge field. This field changes the refractive index of the crystal via the Pockels effect, producing a phase grating. In crystals with the diffusion-driven charge transport, the $\pi/2$-phase shift between this grating and the light fringes results in the direct energy transfer from the pumping beam to the signal beam. The gain factor is the measure of such an exchange:

$$\Gamma = \frac{1}{\ell}\ln\frac{I'_S I_P}{I_S I'_P}. \tag{1}$$

It is expressed via the grating thickness ℓ and the beams intensities before the interaction (I_S, I_P) and after it (I'_S, I'_P). As a parameter of the material, the gain factor [13]

$$\Gamma = \frac{2\pi}{\lambda}\frac{\cos\delta_P}{\cos\theta_S} n_S n_P^2 (\mathbf{e}_S\cdot\mathbf{e}_P)\xi r_{\text{eff}} E^i_{\text{sc}}, \tag{2}$$

depends linearly on the effective Pockels coefficient r_{eff}, which allows one to determine both its value and its sign. The gain factor is also influenced by such optical properties of the crystal as the refractive index n, direction $\mathbf{e}$ of the electric field of the beam, angle θ between the beam direction and the normal to the crystal surface, and angle δ between the wave vector and the ray one. The subscripts S and P mark the signal and pumping beams, respectively.

The factor ξ takes a possible residual domain structure of the sample into account, which depends on the imperfect polishing, possible bipolar photoconductivity, *etc.* If these factors are known, as well as the wavelength λ and the imaginary part of the space charge field E^i_{sc}, the measurement of the gain factor using (1) allows estimating the effective Pockels coefficient

$$r_{\text{eff}} = \mathbf{d}_S(\hat{r}\boldsymbol{\kappa})\mathbf{d}_P, \tag{3}$$

which is a linear combination of components of the Pockels tensor $\hat{r}$. It depends on the beam polarization direction $\mathbf{d}$ (the electric displacement direction of the beam), and the direction $\boldsymbol{\kappa}$ of the grating, and the space charge field.

A measurement of the sample factor ξ is difficult, because it depends on the field direction $\boldsymbol{\kappa}$ as well. This factor can be excluded from the expression for gain factor (2) by means of the method of relative measurements [10].

The geometries should be found, where the identical photorefractive gratings are recorded by two sets of light beams with different polarizations. So, it becomes possible to find a ratio of the effective Pockels coefficients from measurements of the gain factors excluding the sample factor ξ, which is identical in two cases.

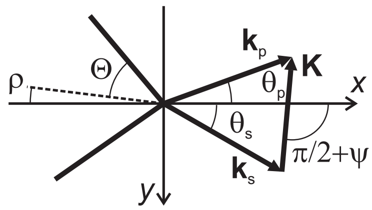


Fig. 1. Orientation of the grating vector K and the wave vectors k_S, k_P in the crystal rotated at an angle ρ

A set of components of the Pockels tensor which form the effective Pockels coefficient depends on a recording geometry. The Pockels tensor for a crystal with one mirror plane xz and the point group m is

$$\hat{r} = \begin{pmatrix} r_{11} & 0 & r_{13} \\ r_{21} & 0 & r_{23} \\ r_{31} & 0 & r_{33} \\ 0 & r_{42} & 0 \\ r_{51} & 0 & r_{53} \\ 0 & r_{62} & 0 \end{pmatrix}. \tag{4}$$

The Pockels effect is indirectly influenced by elastic, acoustooptic, and piezoelectric properties of a crystal [14]. Their common effect is called the secondary Pockels effect. Its magnitude depends on those experimental conditions, which impose restrictions on an elastic deformation. When a photorefractive grating is recorded, a periodic space charge field appears. Its spatial inhomogeneity leads to a complex dependence of the secondary Pockels effect on its direction [15]. The simulation of this dependence for the SPS crystal is impossible at present due to the incomplete data on its acoustooptic tensor [16]. The assumption for the Pockels tensor (4) to be independent of the photorefractive grating direction $\boldsymbol{\kappa}$ is used in the calculation that follows and is discussed in Section 4.

To reveal the r_{42} or r_{62} component in the effective Pockels coefficient, a photorefractive grating must have the y-component. The bigger the value of the y-component, the better becomes the accuracy of the estimate of tensor components. In addition, beam's polarization directions must have both the y- and x-components in the case of r_{62} or both the y- and z-components in the case of r_{42}.

These requirements are fulfilled when a transmission grating is recorded in the x- and z-cut samples, respectively. A geometry, where the signal and the pumping waves impinge on the sample at its x-surface, by making the angle 2Θ in air, is shown in Fig. 1. Let these waves be the crystal eigenwaves with close polarizations. They don't interact if they impinge on the sample symmetrically because of zero effective Pockels coefficient. When the crystal is tilted in the xy-plane (keeping the z-axis direction constant), the orientation of the space charge field and the eigenwave polarizations are varied, which results in the energy exchange between the beams.

Let us consider a change of all parameters that affect the gain factor (2), when the crystal is rotated at an angle ρ. In the initial position, the grating vector direction

$$\boldsymbol{\kappa}_X = (\psi,\, 1,\, 0) \tag{5}$$

is parallel to the y-axis, but the crystal rotation increases its x-component ψ. Using Snell's law and neglecting the dependence of the refractive index on small deviation angles θ of the beams inside the crystal, the grating vector tilt ψ is expressed as

$$\psi = n^{-1}\cos\Theta\sin\rho. \tag{6}$$

The eigenvector of polarization $\mathbf{d}$ of a beam inside the SPS crystal with the wave vector $\mathbf{k}$ is deduced from the wave equation for an anisotropic medium $[\mathbf{s}, [\mathbf{s}, \hat{B}\mathbf{d}] + \mathbf{d}n^{-2} = 0$, where $\hat{B}$ is its inverse dielectric tensor at an optical frequency. In the used coordinate frame, this tensor is represented in the form

$$\hat{B} = \begin{pmatrix} \left(\dfrac{\cos u}{n_1}\right)^2 + \left(\dfrac{\sin u}{n_3}\right)^2 & 0 & \dfrac{1}{2}\left(\dfrac{1}{n_3^2} - \dfrac{1}{n_1^2}\right)\sin 2u \\ 0 & \dfrac{1}{n_2^2} & 0 \\ \dfrac{1}{2}\left(\dfrac{1}{n_3^2} - \dfrac{1}{n_1^2}\right)\sin 2u & 0 & \left(\dfrac{\sin u}{n_1}\right)^2 + \left(\dfrac{\cos u}{n_3}\right)^2 \end{pmatrix}, \tag{7}$$

where $n_1 = 3.0253$, $n_2 = 2.9309$, and $n_3 = 3.0974$ are the principal refractive indices of the SPS crystal at the wavelength $\lambda = 633$ nm [17]. The principal axes of the SPS crystal indicatrix are tilted in the xz-plane at $u = -47.4°$ with respect to the coordinate axes.

When the angle θ of the beam deviation from the normal is small, an approximate solution of the wave equation has the form

$$\mathbf{d}_{XH} = (-\theta,\, 1,\, m_X\theta), \tag{8}$$
$$\mathbf{d}_{XV} = (0,\, m_X\theta,\, 1), \tag{9}$$

where the indices H and V mark the eigenwaves with roughly horizontal and vertical polarizations, respectively. The parameter

$$m_X = \frac{\tan^2 U \sin u \cos u}{1 + \tan^2 U \cos^2 u} \tag{10}$$

depends on the half-angle U between the optical axes of the SPS crystal. It makes 0.265 assuming $U = 39.96°$ at $\lambda = 633$ nm [17]. Using (3), (5), (6), (8), and (9) and Snell's law, the effective Pockels coefficients for the cases of an interaction of both horizontally and vertically polarized beams are as follows:

$$r_{XH} = \sqrt{B_{22}}(r_{21} + 2r_{62} - 2m_X r_{42})\cos\Theta\sin\rho, \tag{11}$$
$$r_{XV} = \sqrt{B_{33}}(r_{31} + 2m_X r_{42})\cos\Theta\sin\rho. \tag{12}$$

Thus, the effective Pockels coefficient is proportional to the $\sin\rho$ (or to the tilt ψ of the grating vector to the y-axis), and the proportionality coefficient includes the desired components of the Pockels tensor.

Other parameters in (2) are assumed to be orientation-independent because of the following reasons. We have $\cos\theta \approx 1$ and $\cos\delta \approx 1$, because the arguments of these factors are small. In view of the crystal symmetry, the refractive index and the factor $(\mathbf{e}_S \cdot \mathbf{e}_P)$ are even functions of the crystal tilt ρ in the considered geometry. Neglecting the second and higher terms of a series, the values $n_H \approx B_{22}$, $(\mathbf{e}_S \cdot \mathbf{e}_P)_H \approx 1 - 2B_{22}\sin^2\Theta$ are obtained for the horizontally polarized eigenwave, and the values $n_V \approx B_{33}$, $(\mathbf{e}_S \cdot \mathbf{e}_P)_V \approx 1$ are obtained for the vertically polarized eigenwave. There, B_{ij} is the respective component of the inverse dielectric tensor (7) of SPS.

The diffusion transport dominates in SPS crystals. The effects of charge screening could be neglected, when the grating spatial frequency K is much smaller than l_s^{-1}, where l_s is the Debye screening length of the material. In this case, the space charge amplitude is proportional to the grating spatial frequency [12]. It decreases, when the crystal is rotated

$$E^i_{\rm sc} \propto K = 4\pi\lambda^{-1}\sin\Theta\cos\rho. \tag{13}$$

Consequently, the gain factor varies as $\sin 2\rho$ with the crystal rotation. Using the estimates for constant parameters and (2), (11)–(13), the dependence of the ratio of the Pockels tensor components on the gain factors Γ_H and Γ_V of the horizontally and vertically polarized beams is found as

$$\frac{r_{21}+2r_{62}-2m_X r_{42}}{r_{31}+2m_X r_{42}}=\frac{B_{22}}{B_{33}(1-2B_{22}\sin^2\Theta)}\frac{\Gamma_{XH}}{\Gamma_{XV}}. \qquad (14)$$

It is not possible to determine both desired components from this equation. It is possible, however, to obtain a similar second equation for another geometry, where light beams impinge on the z-cut sample and record a grating close to the y-axis:

$$\frac{r_{23}+2r_{42}-2m_Z r_{62}}{r_{13}+2m_Z r_{62}}=\frac{B_{22}}{B_{11}(1-2B_{22}\sin^2\Theta)}\frac{\Gamma_{ZH}}{\Gamma_{ZV}}, \qquad (15)$$

where the parameter

$$m_Z=\frac{\tan^2 U\sin u\cos u}{1+\tan^2 U\sin^2 u} \qquad (16)$$

is 0.253 in the case of an SPS crystal. The solution of the system of linear equations (14),(15) in the variables r_{42}/r_{23} and r_{62}/r_{23} is

$$\frac{r_{42}}{r_{23}}=\frac{1-G_Z R_Z+m_Z R(1+G_Z)(1-G_X R_X)}{2m_X m_Z(1+G_X)(1+G_Z)-2}, \qquad (17)$$

$$\frac{r_{42}}{r_{23}}=\frac{R(1-G_X R_X)+m_X(1+G_X)(1-G_Z R_Z)}{2m_X m_Z(1+G_X)(1+G_Z)-2}, \qquad (18)$$

where the symbols $R_X=r_{31}/r_{21}$, $R_Z=r_{13}/r_{23}$, and $R=r_{21}/r_{23}$ are the ratios of known components of the Pockels tensor [9–11]. The symbols G_X and G_Z denote the right-hand sides of Eqs. (14) and (15), which can be determined experimentally.

3. Experiment

To measure the dependence of the gain factor on the SPS crystal rotation angle in the geometries described above, the experimental setup shown in Fig. 2 is used. The linearly polarized beam of a He-Ne laser (with TEM_{00}, 5 mW output power at 633 nm) is divided into 2 beams with a ratio of 1/90 of their intensities. The signal and pumping beams impinge upon the SPS sample, which is mounted on the table rotating about the vertical axis. Due to a small amplification in the used geometries, an attenuation of the pump beaming can be neglected, i.e., the undepleted pump approximation holds. To exclude the light absorption in the sample, the intensities of the signal beam without coupling, I_S, and with it, I'_S, were measured with a photodetector placed beyond the sample.

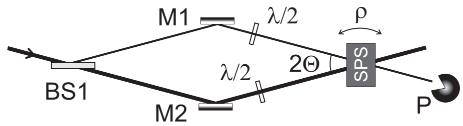


Fig. 2. Experimental setup for measuring the dependence of the gain factor Γ of an SPS crystal on its rotation angle ρ

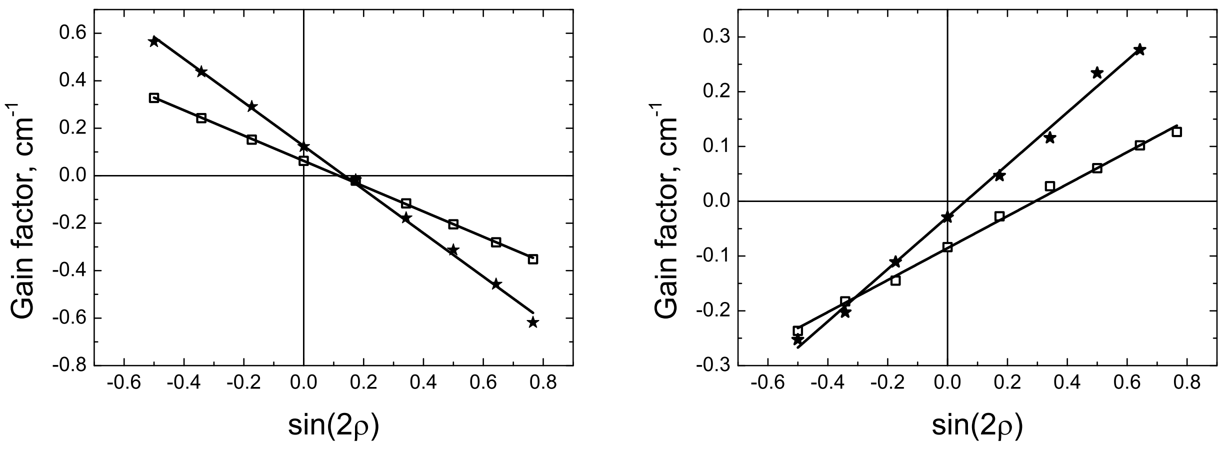
The sample SPS:Te (1%) used for measurements $9\times12\times7$ mm^3 in size was grown at the Uzhgorod National University, Ukraine. It was cut along the crystallographic directions with the accuracy within several degrees of arc. The x- and z-faces of the sample are optically finished. The Te-doped material is chosen deliberately as its gain factor is known to be independent of the light intensity. It was important to exclude the possible effect of the intensity variation, when the sample was rotated.

The signal and pumping beams impinge upon the sample at the angle $2\Theta = 19°$. In this case, the screening effects are reduced, because the grating spatial frequency $K = 4\pi\sin\Theta/\lambda \approx 3.3\ \mu\text{m}^{-1}$ less than the reciprocal Debye length $l_s^{-1} \approx 6\ \mu\text{m}^{-1}$ of the sample [12]. When the signal beam is amplified, its intensity rapidly increases and then slowly decreases. This effect is caused by the formation of two out-of-phase photorefractive gratings in the SPS crystal [1]. The transient peak value of the signal intensity is used to calculate the gain factor according to (1). The angular dependence of the gain factor is shown in Fig. 3, taken at increments of 5° for the geometries of the x- and z-cuts.

In agreement with the calculations presented above, the gain factor depends linearly on the sine of the double value of rotation angle ρ in two geometries.

The intersection point of two lines moved, however, out of the coordinate origin. Its horizontal displacement can be explained by a small tilt (less than 2.5°) of the surface normals to the x- and z-coordinate axes. Its vertical displacement shown in Fig. 3, b is produced by a considerable attenuation of the signal beam transmitted through the z-cut, because of the strong beam fanning in this geometry.

The constant term in the linear dependence of the gain factor on $\sin 2\rho$ does not allow to use formulas (14) and (15) to calculate G_X and G_Z directly. These



a *b*

Fig. 3. Dependence of the gain factor Γ of the SPS crystal on the sine of the double rotation angle $\sin 2\rho$ in the geometries of the x-cut (a) and z-cut (b). Stars and squares represent the results of measurements with the nearly vertically and nearly horizontally polarized eigenwaves of the crystal. Solid lines show the best linear fit of these dependences

equations can be modified, however, in such a way that the slope $\partial\Gamma/\partial \sin 2\rho$ of the angular dependence appears instead of the gain factor itself.

This procedure is well justified, because a deviation of the experimental dependence $\Gamma(\sin 2\rho)$ from a linear function is obviously small, as is seen from Fig. 3. The virtual shift of the coordinate origin doesn't lead to a change of the curve slope, which contains information about the Pockels tensor of the SPS crystal.

4. Discussion

The values $G_X = 0.63 \pm 0.01$ and $G_Z = 0.66 \pm 0.04$ have been calculated using the experimental data shown in Fig. 3. With the known data on $R_X = 1.54 \pm 0.05$, $R_Z = 2.02 \pm 0.08$ [9], and $R = -7 \pm 1$ [10, 11], the desired tensor components have been determined according to (17) and (18) in the form of $r_{42}/r_{23} = 0.26 \pm 0.08$ and $r_{62}/r_{23} = 0.23 \pm 0.12$. Taking the value $r_{21} = 92$ pm/V [7] into account, the non-diagonal components of the Pockels tensor of SPS have been estimated as $r_{42} = -3.4 \pm 1.8$ pm/V and $r_{62} = -3 \pm 2.3$ pm/V. The accuracy of the G_X and G_Z parameters might be improved by increasing the number of experimentally measured points in Fig. 3. However, the main source of errors nests in the imprecise knowledge of R_X, R_Z, and R.

The moduli of the Pockels tensor components ratios obtained above were also calculated, by using the data presented in [10]: $r_{42}/r_{23} = 0.06 \pm 0.01$ and $r_{62}/r_{23} = 0.42 \pm 0.09$. While the estimates for $|r_{62}/r_{23}|$ agree within the error bars, the values for $|r_{42}/r_{23}|$ differ sufficiently.

It should be noted that the measurements of the r_{42} component in [10] were performed with the grating vector directed along the bisector of the y- and z-axes. Thus, the geometry-dependent secondary Pockels effect mentioned above in Section 2 might be a reason for the discrepancy of r_{42} values.

The main advantage of the presented technique of evaluation of a non-diagonal component of Pockels tensor over that based on the photorefractive anisotropic and isotropic Bragg diffraction [10] consists in its sensitivity to the component sign.

The use of the described technique is preferable in case of a week beam coupling. The diffraction efficiency is proportional to r_{eff}^2 and comes down to the noise level faster than the gain factor, which is linearly proportional to r_{eff}. The new technique also gives a better precision in case of a strong beam coupling, when the diffraction efficiency is saturated.

5. Conclusion

A modified technique of determination of the values and the signs of non-diagonal components r_{42} and r_{62} of the Pockels tensor of a monoclinic photorefractive crystal from the self-difraction features is proposed and substantiated. The reliability of the method is experimentally confirmed with the SPS crystal (point group m). The components are estimated to be $r_{42} = -3.4 \pm 1.8$ pm/V and $r_{62} = -3 \pm 2.3$ pm/V.

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ВИМІРЮВАННЯ НЕДІАГОНАЛЬНИХ КОМПОНЕНТ ЕЛЕКТРООПТИЧНОГО ТЕНЗОРА ФОТОРЕФРАКТИВНОГО КРИСТАЛА $Sn_2P_2S_6$

Р е з ю м е

В роботі запропонована методика вимірювання недіагональних компонент r_{42} та r_{62} електрооптичного тензора моноклінного фоторефрактивного кристала. Її було використано для характеризації кристала $Sn_2P_2S_6$.

А.Ю. Волков

ИЗМЕРЕНИЕ НЕДИАГОНАЛЬНЫХ КОМПОНЕНТ ЭЛЕКТРООПТИЧЕСКОГО ТЕНЗОРА ФОТОРЕФРАКТИВНОГО КРИСТАЛЛА $Sn_2P_2S_6$

Р е з ю м е

В работе предложена методика измерения недиагональных компонент r_{42} и r_{62} электрооптического тензора моноклинного фоторефрактивного кристалла. Она использована для характеризации кристалла $Sn_2P_2S_6$.