

# POLARIZATION DEPENDENCE OF TWO-WAVE MIXING IN COUNTERPROPAGATING GEOMETRY IN SILLENITE CRYSTALS

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An investigation of holographic recording in the counterpropagating geometry of two-wave mixing experiments for crystals of the sillenite-group is presented. Three identical samples of  $\text{Bi}_{12}\text{SiO}_{20}$  (BSO),  $\text{Bi}_{12}\text{TiO}_{20}$  (BTO), and  $\text{Bi}_{12}\text{GeO}_{20}$  BGO crystals have been investigated. It was shown that a maximum grating amplitude and a maximum diffraction efficiency are observed for the recording by beams with different directions of linear polarization. The optimal angle between the polarizations of recording beams depends on the value of optical activity and the crystal length. Also, an analytical solution for the diffraction efficiency for different polarization states as well as numerical calculations based on the analytical solution and experimental data are shown.

with different recording polarizations). We also present a qualitative theoretical model which describes the measured experimental results. The calculated numerical results are in good agreement with the experimental results. The dependence of the angle between the polarization states of the recording beams on the optical activity and the crystal length was investigated, and the optimal angle was found.

## Introduction

The investigated crystals of the sillenite-group BSO, BTO and BGO belong to the point group 23. Therefore, they are optically isotropic as well as optically active and exhibit a linear electrooptic effect. These crystals also exhibit comparable short characteristic recording and decay times, which makes them suitable for dynamic holographic interferometry. A counterpropagating geometry of holographic recording is especially interesting since it provides the extremely high sensitivity to external influences [1]. Due to the strong inherent optical activity, a strict theoretical description of the diffraction process in these crystals is rather complicated. Nevertheless, a number of publications discussed this particular problem and found numerical and analytical solutions [2–4]. Though there are a number of publications dealing with the theoretical description of holographic recording in counterpropagating geometry in sillenite-crystals, there are only few experimental data on this subject [5, 6].

In the present paper, we investigate the diffraction efficiency of these crystals for different recording parameters (i.e. the counterpropagating geometry

## 1. Experimental Setup and Procedure

The experimental setup can be seen in Fig.1. We used a frequency-doubled Nd:YAG Laser 7 with an output power of 200 mW for the recording of a hologram in the counterpropagating geometry. The beam is sent through a beam forming system 6, which removes high spatial frequencies and collimates the beam. A  $\lambda/4$ -plate 5 is used to induce a circular polarization of the initially linear polarized beam. Following that, the beam is split up using a non-polarizing beamsplitter and the polarization state of the recording beams is changed again by two polarizers 4. During all experiments, the polarization state of 4a was kept constant (vertical to the plane of the experimental setup), while the polarization state of the second beam was varied using 4b. The angles  $\theta_1, \theta_2$ , and  $\theta_3$  for polarizers 4b, 4c, and 4a, respectively, were measured with respect to the vertical axis. Both beams are directed to the sample from opposite sides using two mirrors (2 and 3) and intersect inside the photorefractive crystal at an angle of about  $175^\circ$ . In order to detect the diffracted signal, we have used the well-known technique of phase modulation of one of the recordings beams [7]. A high frequency phase modulation (i.e. in comparison with the inverse recording time) was accomplished by a piezo driven mirror 2 which oscillated at about 17 kHz. The light intensity at the crystal output is a result of the interference of the transmitted reference

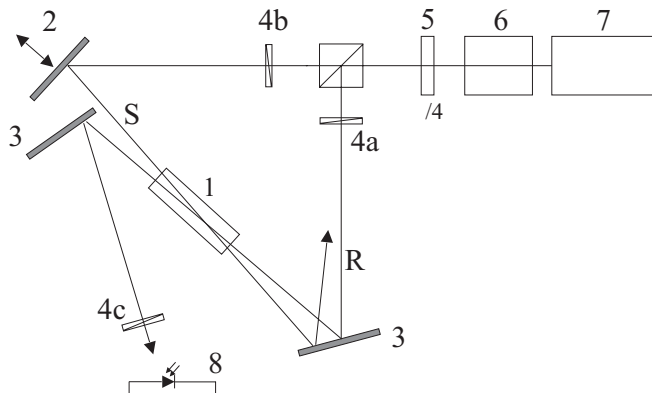


Fig. 1. 1 — photorefractive crystal, 2 — piezo driven mirror, 3 — mirror, 4 — polarizer 5 —  $\lambda/4$ -plate, 6 — pinhole and collimation lens, 7 — Nd:YAG Laser, 8 — photodiode

beam and the phase modulated diffracted signal beam. This light intensity was detected by a photodiode, with polarizer 4c installed in front of it. Using this additional polarizer, it is possible to detect the difference in the polarization states of the transmitted reference beam and the diffracted signal beam. Since the amplitude of the detected signal is proportional to the interference of the transmitted and diffracted beams, the strongest signal should be detected for the case of equal projection amplitudes (on the polarizer axis) for both beams (see Fig. 2). Due to strong instabilities of the first harmonic of the signal, we measured the oscillation amplitude of the light intensity at the second harmonic of the phase modulation with a selective nanovoltmeter (i.e. the nanovoltmeter can be tuned to detect only certain frequencies).

## 2. Experimental Results

During our experiments, we investigated three different crystals of the selenite group, namely BSO, BTO and BGO. All samples were  $5 \times 2 \times 3$  mm in size (these values correspond to the  $[111]$ ,  $[1\bar{1}\bar{1}]$  and  $[1\bar{1}0]$  crystallographic axes, respectively). The recording beams were propagating along the  $[111]$  axis which is parallel to the long edge of the crystal (cf. Fig. 4). The dependence of the signal amplitude on the polarization of the recording beams, as well as the orientation of the polarizer in front of the photodiode was measured. Due to the optical activity, the effective interaction area inside the crystal changed with the variation of the polarization of the recording beams. It

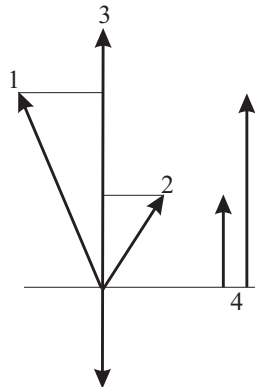


Fig. 2. Polarization states before detection: 1 — amplitude of transmitted beam, 2 — amplitude of diffracted beam, 3 — angular position of polarizer (4c), 4 — projection of (1) and (2) onto the polarizer axis

also depended on the value of optical activity and length of the crystal. In the present paper, we concentrate on optimizing the polarization states of the recording beams in order to obtain the maximum amplitude of the detected signal. These conditions will correspond to the optimum configuration for a two-wave mixing experiment in the counterpropagating geometry. Fig. 3 shows the measured signal amplitudes as a function of the angles  $\theta_1$  and  $\theta_2$ . The latter angle was changes during the experiments from 0 to  $180^\circ$ , while  $\theta_1$  was changed from 0 to  $90^\circ$ . In Fig. 3,b, a periodicity of  $180^\circ$  can be found along the  $\theta_2$  axis. Also, the maximum of the detected signal for the crystals with different values of the optical activity (BTO — 12 deg/mm, BGO — 37.2 deg/mm, and BSO — 38 deg/mm [8]) was observed at substantially different angles between the polarizations of the recording beams. We have developed a theoretical model which describes the experimental results and allows one to predict the optimal condition for a two-wave mixing setup.

## 3. Theoretical Description

### 3.1. Formation of the Interference Pattern

First of all, we consider the formation of the interference pattern by two recording beams. Since the diffraction efficiency is rather low, we neglect the effect of two-wave mixing (the change of the intensities of the recording beams due to diffraction). However, we take into account the change of the states of polarization of the recording beams due to optical activity, which influences the contrast of the interference pattern. In the case of the

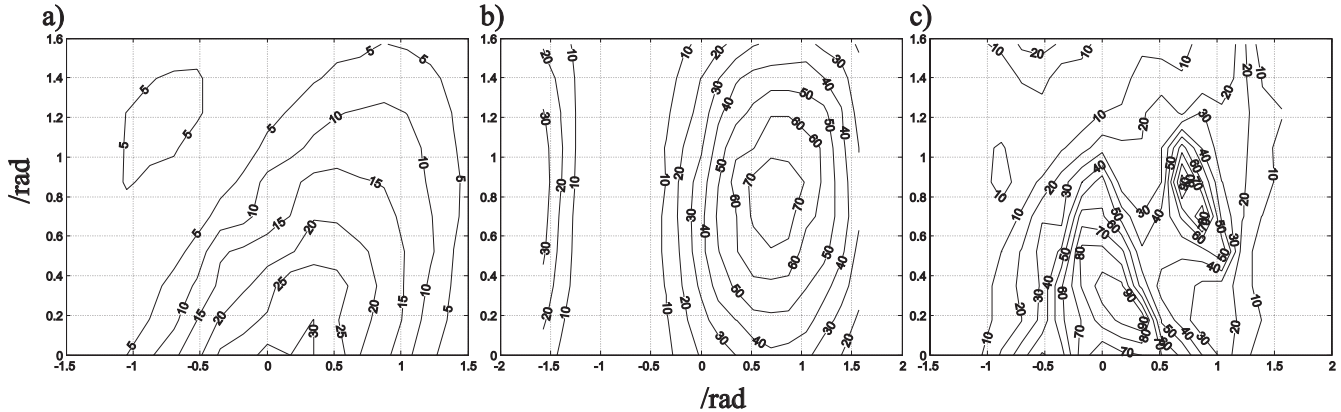


Fig. 3. Contour plot of experimental results: signal amplitude in arbitrary units as a function of angles  $\theta_1$  and  $\theta_2$ . a – BGO, b – BTO, c – BSO

counterpropagating geometry, both recording beams propagate along one axis (in our case the crystallographic axis [111], see Fig. 4) in opposite directions and amplitude variations of the reference ( $R$ ) and signal ( $S$ ) beams can be represented as follows:

$$R_x(z) = a\sqrt{I_0} \exp\left(-\frac{\alpha}{2}z\right) \cos \rho z,$$

$$R_y(z) = a\sqrt{I_0} \exp\left(-\frac{\alpha}{2}z\right) \sin \rho z,$$

$$S_x(z) = (1-a)\sqrt{I_0} \exp\left(\frac{\alpha}{2}(z-l)\right) \cos(\theta_1 + \rho(z-l)),$$

$$S_y(z) = (1-a)\sqrt{I_0} \exp\left(\frac{\alpha}{2}(z-l)\right) \sin(\theta_1 + \rho(z-l)), \quad (1)$$

where  $a$  is the coefficient of the beam splitter ( $a = 0.6$  in our case),  $\alpha$  is the absorption coefficient,  $\rho$  is the coefficient which characterizes the present optical activity (38 deg/mm for BSO, 37.2 deg/mm for BGO, 12 deg/mm for BTO at 532 nm [8]) in the crystal.  $I_0$  is the total intensity of the recording light,  $\theta_1$  is the angle between the linear input polarization of the signal beam ( $S$ ) and the polarization of the reference beam ( $R$ ) (or the  $x$  axis, respectively), and  $l$  is the crystal length. The contrast of the interference can be described by the following expression:

$$m(\theta_1, z) = 2 \frac{R_x S_x + R_y S_y}{|R|^2 + |S|^2}. \quad (2)$$

Substituting Eqs. (1) into (2) yields the following expression for the contrast of the interference pattern, based on the optical activity of the crystal and the initial polarization states:

$$m(\theta_1, z) =$$

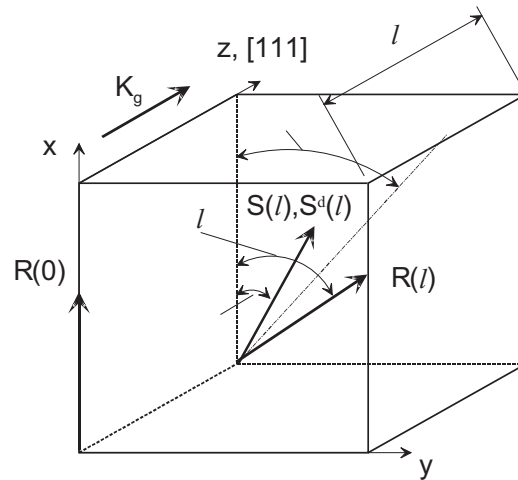


Fig. 4. Two-wave mixing experiment in the counterpropagating geometry considering optical activity

$$= \frac{2a(1-a) \exp\left(-\frac{\alpha}{2}l\right)}{a^2 \exp(-\alpha z) + (1-a)^2 \exp(\alpha(z-l))} \cos(\rho l - \theta_1). \quad (3)$$

### 3.2. Diffraction

In the following section, we will briefly discuss the formation of a grating and the diffraction of the readout beam therefrom. In the considered case of counterpropagating beams, the grating wave vector  $K_g$  is directed along [111]. In this particular geometry the grating amplitude does not depend on the direction of polarization of the readout beam ( $S$ ). Using the matrix formalism for the calculation of the propagating

beams (and considering the linear electrooptic effect), the grating amplitude can be calculated as follows [5].

$$n_1 = \frac{1}{2\sqrt{3}} n_0^3 r_{14} E_{SC}, \quad (4)$$

where  $n_0$  is the average refractive index of the crystal,  $r_{14}$  is the electrooptic coefficient and  $E_{SC}$  is the space charge field which is proportional to the contrast of the interference pattern,

$$E_{SC} \sim im(\theta_1, z). \quad (5)$$

The imaginary coefficient  $i$  describes the  $\pi/2$  shift of the refractive index grating with respect to the interference pattern, which is due to the diffusion-dominated transport mechanism during the recording process. In order to describe the diffraction process, it is necessary to consider the optical activity of sillenite crystals. The linear polarization of the readout beam (signal beam  $S$ , see Fig. 4 and 1) is rotated during propagation through the crystal. However, since the grating amplitude is independent of the polarization, the optical activity does not influence the process of diffraction in the used geometry. Furthermore, the readout beam and the corresponding diffracted beam (i.e. after reflection from the grating) have the same state of polarization at any point inside the crystal because their polarization is rotated about the same amount inside the crystal but in the opposite direction during propagation. Thus, the polarization state after diffraction from the grating returns to its initial state as it was before entering the crystal (a linear polarization tilted about  $\theta_1$  with respect to the vertical axis). In this case, the diffraction process of the signal beam  $S$  in the low diffraction efficiency approximation (we do not consider a change of the amplitude due to diffraction) can be described by the following scalar differential equation.

$$\frac{d|S^d|}{dz} \sim m(\theta_1, z) r_{14} |S(l)|. \quad (6)$$

Substituting (1) and (3) into (6) yields

$$\begin{aligned} \frac{d|S^d|}{dz} &\sim 2a(1-a)^2 \sqrt{I_0} r_{14} \cos(\rho l - \theta_1) \times \\ &\times \frac{\exp(-\alpha l) \exp(-\frac{\alpha}{2} z)}{a^2 \exp(-\alpha z) + (1-a)^2 \exp(\alpha(z-1))}. \end{aligned} \quad (7)$$

Since the absorption of the crystal does not influence the dependence of the signal of the diffracted beam on the polarization, but rather reduces the amplitude of the

signal, the effect of absorption will not be considered in the theory at the moment. However, it should be kept in mind, that absorption should be included in various normalizing coefficients. Solving the differential equation (7) yields a simple formula for the amplitude of the signal beam:

$$|S^d(l)| \sim 2 \frac{a(1-a)^2}{a^2 + (1-a)^2} \sqrt{I_0} r_{14} \cos(\rho l - \theta_1) l. \quad (8)$$

### 3.3. Signal Detection

In our experiments, we detected the AC photo current at the second harmonic of phase modulation of the signal beams  $S$ . The oscillating component of the light intensity detected by the photodetector is generated by the interference of one transmitted and the diffracted signal beam. An installed polarizer in front of the detector was used to extract the information about the polarization difference of both beams. The angle  $\theta_2$  was measured between the axis of the polarizer and the vertical  $x$  axis. Thus, in the experiment, the polarization of both beam was projected onto the axis of the polarizer. This effect has to be taken into account when calculating the resulting signal amplitude (or the dependence thereof on the polarization states  $\theta_1$  and  $\theta_2$ ). The scalar amplitude of both interfering beams is:

$$R_{\theta_2}(l) = |R(l)| \cos(\rho l - \theta_2), \quad (9)$$

$$S_{\theta_2}^d(l) = |S^d(l)| \cos(\theta_2 - \theta_1) \exp(i\delta \cos \Omega t), \quad (10)$$

where  $R(l) = a\sqrt{I_0}$  is the modulus of the amplitude of the reference beam after leaving the crystal (in the low diffraction efficiency approximation and without absorption),  $S^d(l)$  is the amplitude of the diffracted beam, and  $\cos(\rho l - \theta_2)$  is the projection of the polarization of the reference beam onto the polarizer axis taking into account the optical activity in the crystal. The term  $\cos(\theta_2 - \theta_1)$  describes the projection of the diffracted beam polarization onto the polarizer axis,  $\delta$  is the amplitude of the phase modulation while  $\Omega$  is the frequency of the phase modulation. Taking all previous considerations into account and for a weak phase modulation  $\delta \ll 1$ , the dependence of the detected signal amplitude as a function of the polarization states of both recording beams and the analyzer in front of the detector can be calculated as follows:

$$\begin{aligned} I_{2\Omega}(\theta_1, \theta_2) &\sim R_{\theta_2}(l) S_{\theta_2}^d \sim \delta^2 \frac{a^2(1-a)^2}{a^2 + (1-a)^2} r_{14} I_0 l \times \\ &\times \cos(\rho l - \theta_1) \cos(\rho l - \theta_2) \cos(\theta_2 - \theta_1). \end{aligned} \quad (11)$$

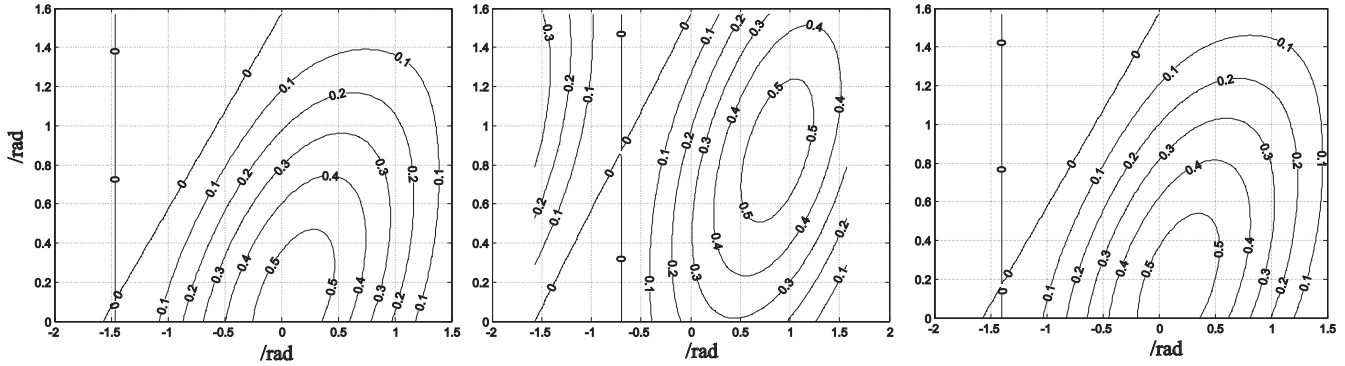


Fig. 5. Calculated contour-plot for the BGO, BTO and BSO crystal, horizontal and vertical axis indicate angles  $\theta_2$  and  $\theta_1$

#### 4. Numerical Calculation

In this section, we present numerical calculations based on the theoretical results of the previous section. We concentrate on the optimization of the detected signal in order to find an optimal configuration for two-wave experiments. Therefore, we calculated the normalized dependence of the signal on the polarization states of the recording beams and compared the results with our experimental data. The three contour plots (Fig. 5) for the used sillenite crystals BSO, BTO, and BGO were calculated for  $\rho = 38$  deg/mm,  $\rho = 10$  deg/mm, and  $\rho = 37.2$  deg/mm, respectively. The length  $l$  we used in the simulation for the crystals was 5 mm. Apparently the simulations reproduce our measured experimental results rather well (cf. Fig. 5 and 3). The contour plot of the theoretical signal amplitude shows an elliptical symmetry which is due to the dominant three cosine terms (see Eq. (11)) and can also be found in the experimental data. As can be seen from the figures, the position of the signal maximum strongly depends on the type of crystal. Corresponding to Eq. (11), the optical activity (besides the length of the crystal) is the only parameter that influences the position of the signal maximum. Our experimental results and theoretical calculations show that, for a given crystal length of 5 mm, the maximum two-wave mixing in the counterpropagating geometry will be observed in a rather non-trivial configuration of the experiment: the holographic recording is performed by beams with different linear polarizations, and the angle between the polarizations of the recording beams depends on the crystal optical activity.

#### Discussion

An investigation of holographic recording in the counterpropagating geometry of two-wave mixing experiments for crystals of the sillenite group has been presented. During our experiments, we investigated three identical samples of BSO, BTO and BGO crystals. It is shown that the maximum grating amplitude and maximum diffraction efficiency are observed for recording by beams with different yet very specific linear polarizations. A thorough theoretical analysis has been carried out, as presented in Section 4. We find a good agreement between the simulations and the experimental data for the BGO crystal and at least a qualitative description for the BTO and BSO crystals. The obvious tilt in the theoretical simulation of the BTO crystal in comparison with the experimental data most probably stems from an yet uninvestigated parameter during the measurement. Our experiments have also shown that the optimum angle between the polarizations of the recording beams depends on the value of the optical activity and the crystal length. The results of these investigations can be used for the optimization of two-wave mixing setups using this specific geometry. Due to the considerable low grating period (down to 100 nm for our particular setup), the investigated geometry of counterpropagating recording beams is very promising for adaptive interferometers based on reflection dynamic holograms.

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ПОЛЯРИЗАЦІЙНА ЗАЛЕЖНІСТЬ ЗУСТРІЧНОЇ  
ДВОПУЧКОВОЇ ВЗАЄМОДІЇ  
В КРИСТАЛАХ СИЛЕНІТІВ

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Резюме

Розглянуто голографічний запис в кристалах групи силенітів двома пучками світла, що поширюються в зустрічних напрямках. Досліджено три подібні зразки кристалів BSO, ВТО та ВГО. Показано, що максимальна амплітуда ґратки та максимальна дифракційна здатність спостерігаються для пучків з різним нахилом площини лінійної поляризації, який залежить від сталої оптичної активності та від товщини зразка. Наведено аналітичний вираз для залежності дифракційної здатності ґратки від поляризації хвиль, що її записують. Чисельні розрахунки порівнюються з результатами експерименту.