Coupling of counterpropagating light beams in low-symmetry crystals

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Received April 10, 2009; accepted May 27, 2009;
posted June 9, 2009 (Doc. ID 109992); published July 7, 2009

Two-beam coupling gain enhancement is demonstrated for a nontraditional orientation of counterpropagating waves in the monoclinic crystal $Sn_2P_2S_6$. © 2009 Optical Society of America
OCIS codes: 190.5330, 160.2260.

Tin hypothiodiphosphate ($Sn_2P_2S_6$, SPS) is a promising nonlinear material that combines a relatively high two-beam coupling gain (of the order of 10 cm^{-1}) with quite a fast response (in millisecond range) [1]. Another attractive feature of this material is its sensitivity to near IR radiation that extends to optical communication wavelengths of up to 1.5 μm [2]. However, the great potential of this material has still not been explored in full: the largest two-beam coupling gain factor of 38 cm^{-1} reported for the so called "brown SPS" in [1] is still smaller than that expected for a crystal with such a high electro-optic coefficient of $r_{111} \simeq 174$ pm/V [3].

The reduction in the two-beam coupling gain is caused by a limitation of the space charge field. This follows directly from the well known expression for the gain factor [4]

$$\Gamma = \left(\frac{2\pi K n^3 r_{\text{eff}}}{\lambda}\right)\left(\frac{k_B T}{e}\right)\left(\frac{1}{1+\ell_s^2 K^2}\right), \qquad (1)$$

where $K = 2\pi/\Lambda$ is the spatial frequency of the photorefractive grating; λ is the light wavelength in vacuum, $\Lambda = \lambda/(2n \sin \theta)$ is the grating spacing; n is the index of refraction; r_{eff} is the effective electro-optic coefficient; k_B is the Boltzmann constant; T is the absolute temperature; e is the electron charge; and ℓ_s is the Debye screening length, $\ell_s = \sqrt{(\epsilon\epsilon_0/N_{\text{eff}})(k_B T/e)}$, with the effective trap density $N_{\text{eff}} = (N_D - N_D^+)N_D^+/N_D$, and N_D, N_D^+ represent the total density of donors and the ionized donor density, respectively.

At ambient temperature SPS is close to the ferroelectric phase transition (Curie temperature of 339 K [1]) and the relevant dielectric permittivity is large enough, $\epsilon_{11} \simeq 230$, to make the Debye screening length rather large. This in turn reduces Γ owing to the denominator of the third factor in Eq. (1). This limitation on the space charge field becomes especially severe for coupling between counterpropagating beams, for which the grating spacing is the smallest possible, $\Lambda = \lambda/2n$. Here the inequality $(\ell_s K)^2 \gg 1$ holds and Eq. (1) can be rewritten as

$$\Gamma = \left(\frac{2\pi^2 n^4}{\lambda^2}\right)\left(\frac{N_{\text{eff}} r_{\text{eff}}}{\epsilon\epsilon_0}\right). \qquad (2)$$

The usual way to mitigate space charge limitations is by using technological modifications to the crystal. Intentional doping or aftergrowth treatment may increase N_{eff}, making the Debye screening length smaller and thereby increase the space charge field at a given spatial frequency of the recorded index grating (see, e.g., [1,5]). For the strong screening in the case of the reflection gratings, the gain factor should depend linearly on the trap density [see Eq. (2)].

In this Letter an alternative method of two-beam coupling enhancement is considered, which consists of finding an optimum orientation of the interacting waves with respect to the crystal axes, while profiting from the strong anisotropy of the dielectric properties and electro-optic properties of SPS. We are searching for particular orientations of the grating vectors $\mathbf{K}$, for which the reduction in the gain factor through a smaller effective electro-optic coefficient is largely overcompensated by an increase in the space charge field arising from reduced screening (smaller dielectric constant). Special attention is given to the interaction of counterpropagating waves.

Simple as it may appear, this technique is not trivial for crystals of low symmetry, for example, for crystals of monoclinic m class; the optical frame here coincides neither with the crystallographic frame (where the electro-optic tensor is defined) nor with the dielectric frame [6,7]. In addition, the available data on dielectric properties of SPS are rather limited [8,9] and quite contradictory. Referring to publication [8] we present the ratio of permittivity tensor components normalized to ϵ_{xx} as $\epsilon_{xx}:\epsilon_{yy}:\epsilon_{zz}:\epsilon_{xz} \simeq 1:0.1:0.25:0.14$ [10]. In part these data have been confirmed by purely optical measurements [11] that gave $\epsilon_{xx}:\epsilon_{zz} \simeq 1:0.25$.

From the above permittivity tensor data $\hat{\epsilon}$, we can expect the smallest screening to be for a space charge grating with the grating vector aligned along the y axis. Unfortunately, the Pockels tensor of SPS

0146-9592/09/142126-3/$15.00

$$\hat{r} = \begin{pmatrix} r_{xxx} & 0 & r_{xxz} \\ r_{yyx} & 0 & r_{yyz} \\ r_{zzx} & 0 & r_{zzz} \\ 0 & r_{yzy} & 0 \\ r_{xzx} & 0 & r_{zxz} \\ 0 & r_{yxy} & 0 \end{pmatrix} \tag{3}$$

does not possess nonvanishing electro-optic coefficients with identical first two indices for a space charge field aligned along the y axis. Therefore a grating recording with the two identically polarized eigenwaves and $\mathbf{K}\|\mathbf{y}$ is impossible. So the intention here is to tilt the grating vector $\mathbf{K}$ to a certain angle β in the xy plane to make use of the largest Pockels tensor component r_{xxx} [3] while still keeping the permittivity relatively small.

To estimate the expected gain enhancement, it is necessary to know the tilt angle dependences of the effective electro-optic coefficient $r_{\text{eff}}(\beta)$, the effective dielectric permittivity $\epsilon_{\text{eff}}(\beta)$, and the indices of refraction $n_{2,3}(\beta)$ that appear in Eq. (2). The angular dependences of the gain factor for two eigenpolarizations, calculated with all the above factors taken into account and with the hierarchy of electro-optic coefficients from [6], are shown in Fig. 1. The gain factor here is normalized to its value for the signal wave polarized along the z axis and $\mathbf{K}$ aligned along the x axis.

The simplest of the three mentioned dependences is that for the effective dielectric permittivity in xy plane,

$$\epsilon_{\text{eff}} = \epsilon_{yy} \sin^2 \beta + \epsilon_{xx} \cos^2 \beta. \tag{4}$$

More complicated expressions for the effective electro-optic coefficients follow from the structure of the Pockels tensor [Eq. (3)]. Below we present the dependences for two eigenwaves of the xy crystallographic plane. For $\beta=0°$ the polarization vectors of these eigenwaves are perpendicular or parallel to the xy plane, with appropriate superscripts used to differentiate between the two cases,

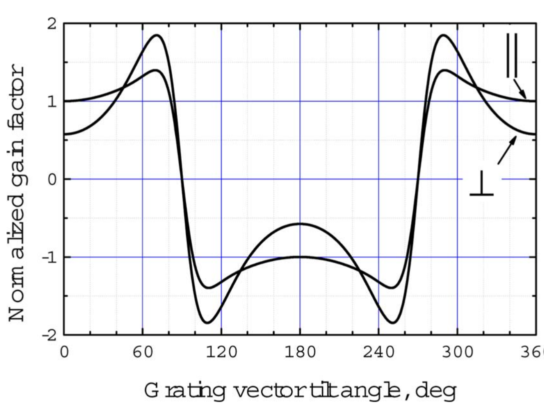

Fig. 1. (Color online) Calculated dependence of the gain factor on direction angle of grating vector $\mathbf{K}$ in the xy plane and eigenpolarizations of the recording waves.

$$\begin{aligned} r_{\text{eff}}^{\|} = {} & (r_{xxx} - 2r_{yxy})\sin^2 \beta \cos \beta \cos^2 \nu \\ & + r_{yyx} \cos^3 \beta \cos^2 \nu + r_{zzx} \cos \beta \sin^2 \nu \\ & - 0.5(r_{xzx} - r_{yzy})\sin 2\beta \sin 2\nu, \end{aligned} \tag{5}$$

$$\begin{aligned} r_{\text{eff}}^{\perp} = {} & (r_{xxx} - 2r_{yxy})\sin^2 \beta \cos \beta \sin^2 \nu \\ & + r_{yyx} \cos^3 \beta \sin^2 \nu + r_{zzx} \cos \beta \cos^2 \nu \\ & - 0.5(r_{xzx} - r_{yzy})\sin 2\beta \sin 2\nu. \end{aligned} \tag{6}$$

It should be noted that the eigenwave polarization does not remain the same when β changes because the optical indicatrix of SPS is tilted roughly to 45° with respect to the x and the z axes. These changes are accounted for in Eqs. (5) and (6) by the angle ν that the electric field of the light wave makes with respect to the xy plane; the equation that interrelates ν and β is given in [11]. In general, the index of refraction that appears in Eq. (2) is also β dependent, but these changes cannot qualitatively affect the dependencies of Fig. 1 and they are therefore neglected.

As one can see the gain factor has certain finite values for a grating vector $\mathbf{K}\|\mathbf{x}$ ($\beta=0°$, 180°, and 360°) but vanishes if $\mathbf{K}\|\mathbf{y}$ ($\beta=90°$, 270°). It is also obvious that the largest value of the gain is achieved anywhere in between the crystal x and y axes. Depending on the polarization, enhancements of the gain factor from 1.25 to 3 times can be expected if the grating vector is turned to ≈20° from the crystal y axis.

In our experiment the beam coupling was studied using SPS:Te (1 wt. %) [5] with the setup shown schematically in Fig. 2. The sample measuring 8 mm $\times$ 4 mm $\times$ 6.5 mm was cut along the crystallographic axes, with optically finished x and y faces. The linearly polarized output of a 30 mW TEM_{00} He–Ne laser operating at 633 nm was used. The laser beam

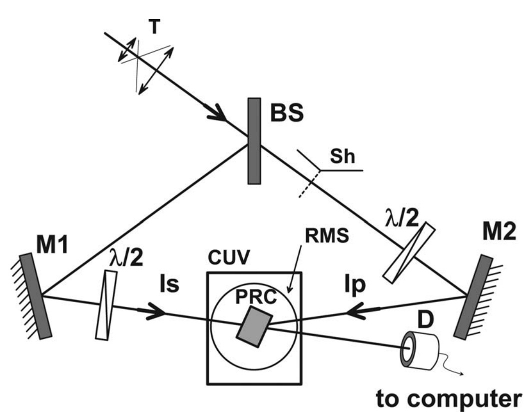

Fig. 2. Experimental setup for gain measurement. With a beam splitter (BS), the two recording beams, signal and pump, are formed from the He–Ne laser beam, which is expanded with a telescope (T). The PRC is placed into the CUV filled with the index-matching oil. Its angle with respect to the recording waves can be adjusted with the RMS. Two phase retarders $\lambda/2$ are used to adjust the eigenpolarization of the recording waves for any particular sample tilt angle. The shutter (Sh) is used to open and close the pump beam; the transmitted signal beam intensity is measured with the detector (D).

was expanded with a 3× telescope (T) to prevent any thermal lens development. The pump and the probe waves were slightly misaligned (to 5°) with an intensity ratio of 1000:1.

The sample was mounted on a magnetic holder, which was immersed into a cuvette (CUV) with optical windows and filled with oil to reduce the refractive index mismatch ($n=1.5$). A specially designed rotating magnet stage (RMS) allowed for the control of the sample tilt angle inside the immobile CUV, increasing the beam propagation angle inside the photorefractive crystal (PRC).

We measured the standard two-beam coupling gain factor

$$\Gamma = \frac{1}{\ell}\ln\frac{I_s}{I_s^0} \tag{7}$$

as a function of the sample rotation angle inside the CUV to reconstruct the dependence of Γ on the grating vector angle β. Here I_s and I_s^0 are the intensities of the transmitted signal wave with and without the pump wave present in the sample, respectively, and ℓ is an interaction length.

Half-wave phase retarders ($\lambda/2$) were placed in each of the two interacting waves to adjust the eigenwave polarization for any new sample rotation angle. The measurements were done for two eigenwaves with the indices n_1 and n_3 [1,12]. Figure 3 shows the results for light beams that enter the sample through the [010] faces and the best fits of the calculated dependences to the measured data.

We also measured the gain factor at $\beta=0$ with beams that enter the sample through the [100] faces. For light polarized along the z axis, the gain factor $\Gamma=2\ \mathrm{cm}^{-1}$. Thus a 1.5 times enhancement of the gain factor is achieved for the coupling of counterpropagating waves, which is in qualitative agreement with our expectations. It is known that the Curie temperature for doped SPS differs from that for nominally undoped material; therefore the dielectric permittivity tensor can be considerably affected by the doping. The absence of reliable data on the $\hat{\epsilon}$ tensor for SPS:Te currently prevents us from the quantitative comparison with the result of the calculation.

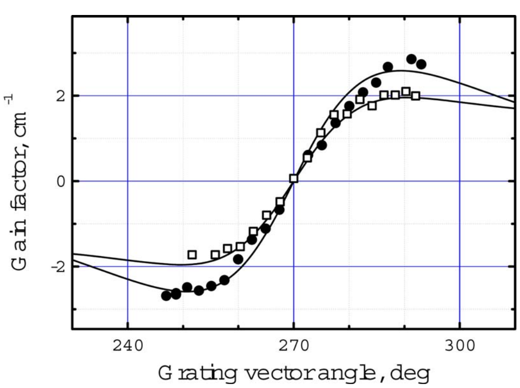

Fig. 3. (Color online) Dependences of the gain factor on the direction angle of grating vector **K** in xy plane and eigenpolarization of the recording waves. Dots and squares show data measured for two eigenpolarizations, perpendicular and parallel, respectively. The solid curves are the best fits of the calculated dependences to the experimental data.

We conclude that the proposed technique proved to be efficient. The absolute value of the improved gain factor is still rather modest, $\Gamma \approx 3\ \mathrm{cm}^{-1}$. However, we can expect that by optimizing the geometry of the interaction a much larger gain can be achieved for SPS crystals, in which the Debye screening length is already reduced by appropriate technological changes to the growth or the poling conditions.

The authors thank A. Grabar and I. Stoyka for SPS samples. A part of this work was conducted during S. Odoulov's stay at the Air Force Research Laboratory. The financial support from the European Office of Research and Development via Science and Technology Center, Ukraine (project P335), is gratefully acknowledged.

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