# OPTIMIZATION OF A RING-LOOP SELF-PUMPED OSCILLATOR BASED ON $\mathrm{Sn_2P_2S_6}$ PHOTOREFRACTIVE CRYSTAL

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The main parameters of a ring self-pumped phase conjugate mirror with the  $\rm Sn_2P_2S_6$  photorefractive crystal are studied in dependence of the experimental geometry factors. A numerical model describing the variation of the phase conjugate reflectivity and dynamic characteristic is proposed. The model can be used for the optimization of the performance of the photorefractive elements based on  $\rm Sn_2P_2S_6$  crystals.

#### Introduction

Phase conjugation (PC) is a process used in applied optics for image processing, laser beam cleanup, etc. [1]. A number of schemes realizing the phase conjugation is described in literature [2—4], and photorefractive (PR) crystals are among the most available media for such a purpose. One of the schemes, the so-called ring-loop self-pumped phase conjugate (SPPC) mirror [5], is one of the most effective and useful ones due to many advantages: the lowest generation threshold among other known SPPC schemes [2—4], as well as no sensitivity to mechanical vibrations and the coherence length of a laser source [6]. The last feature is important for the exploiting with semiconductor lasers. This scheme was realized on the base of several PR crystals: Rh:BaTiO<sub>3</sub> [7, 8], KNbO<sub>3</sub> [6] and other.

Nevertheless, a search of new effective PR materials is still an actual task, especially for the use with sources operating in red and infra-red spectral ranges. One of such new promising materials are  $Sn_2P_2S_6$  crystals exhibiting high enough sensitivity and relative fast (in scale of milliseconds) response [9, 10]. Recent works show a great potential for improving the PR parameters of these crystals by a modification of the crystal growth conditions[11, 12] or goal-directed doping [13]. Another way of improving the crystal performance in PR schemes can be the optimization of a sample orientation relatively to the crystallographic axes.

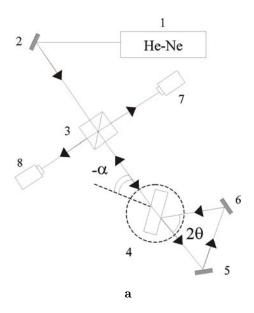
The demonstration and analysis of the wave conjugation specialty in a  $\mathrm{Sn_2P_2S_6}$  crystal used as a

PR medium was done in [14, 15]. A further progress was achieved in [12], where the high performance of the conjugation with the effectiveness up to 40% and the generation time on scale of milliseconds were obtained in nominally pure and modified "brown" crystals. It was shown that the achieved reflectivity is close to a theoretical limit and is restricted by optical losses in the scheme. The best performance conditions were found empirically and no optimizations of the geometric factors including the crystal cut, sample thickness, loop angle, etc., were done. A further improvement of the conjugator requires the optimization of the "geometric" factors. Because Sn<sub>2</sub>P<sub>2</sub>S<sub>6</sub> crystals belong to a monoclinic symmetry (class m), the main axes of the material tensors determining the PR properties are in general case differently oriented in the XZ mirror plane, that complicates the calculations and analysis and requires special studies.

The aim of the present work was to analyze the optimal configuration of the ring-loop SPPC scheme and to build a numerical model allowing one to optimize such a device containing the  $\rm Sn_2P_2S_6$  sample as an active element. For this purpose we studied the experimental dependences of the amplitude and dynamic characteristics of the ring-loop SPPC mirror based on  $\rm Sn_2P_2S_6$  sample on the geometry of the experimental scheme and calculated these parameters on the base of the sample data.

## Experimental Studies of the Ring-loop Self-pumped Phase Conjugate Mirror

The experiments on the phase conjugation in the ringloop scheme were performed on the sample of nominally pure  $Sn_2P_2S_6$  with the dimensions  $7.8\times10.4\times6.0$  mm along X,Y, and Z, respectively. Here, we used the coordinate system convenient for  $Sn_2P_2S_6$ , where the Xaxis is close to the polar direction and Y is the normal to the mirror plane m. The sample was perfectly polished



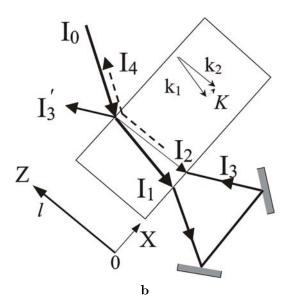


Fig. 1. Experimental scheme for ring-loop self-pumped phase conjugation with  $Sn_2P_2S_6$  crystal. All the beams are polarized in the plane of their propagation. a — experimental setup: 1 — laser, 2, 5, 6 — mirrors, 3 — beam splitter, 4 — sample on the rotating stage, 7, 8 — photodiodes; b — the sample orientation and the used coordinate system. Here, K is the photorefractive grating wave vector, formed by the interference of laser beams with the  $k_1$ ,  $k_2$  wave vectors

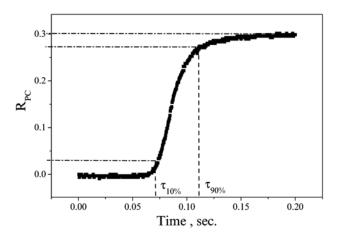


Fig. 2. Time evolution of phase-conjugate reflectivity  $R_{\rm PC}$  after turning on the input beam and definition of the reflectivity rise time (the time between 10 to 90% of the saturated reflectivity)

(Z- and Y-faces) and poled. A He-Ne laser with the light wavelength  $\lambda=633$  nm, max power ~10 mW and Gaussian beam radius of 0.7 mm, was used as a source. All the beams in the experimental scheme (Fig. 1) were polarized in the plane of their propagation. The total reflection of two mirrors which form a ring loop behind the sample was measured to be 0.89. The phase conjugated reflectivity and its time evolution after

the switching on the laser beam by a mechanical shutter were registered by a photodiode and an AD card in a computer.

The PC reflectivity  $R_{PC}$  was determined as the ratio of the PC beam intensity  $(I_4 \text{ in Fig. } 1, b)$  to the incident beam intensity  $I_1$ . Accounting the reflections by the input crystal face, this can be expressed as

$$R_{\rm PC} = \frac{I_{\rm PC}}{I_0 (1 - R_T)^2}.$$
 (1)

The sample face reflection  $R_T$  was calculated by the known Fresnel relations using the measured data on the main values of the refractive indices and the orientation of the optical indicatrix [18].

The beam-conjugate reflectivity was measured as a function of both the loop angle  $2\theta$  and the incidence angle  $\alpha$  of the input beam (Fig. 1,a). The sample rotation in the scheme depicted in Fig. 1,a leads to the simultaneous variations of the azimuth and modulus of the photorefractive grating wave vector K relatively to the crystallographic axes in the XZ plane.

The typical experimental dependence of the PC reflectivity time evolution, calculated according to (1), is shown in Fig. 2. The PC wave build-up time was determined as the time interval between 10 and 90% of saturated PC wave intensity ( $I_4$  in Fig. 1,b) analogously to other works [8, 12], because this time interval depends

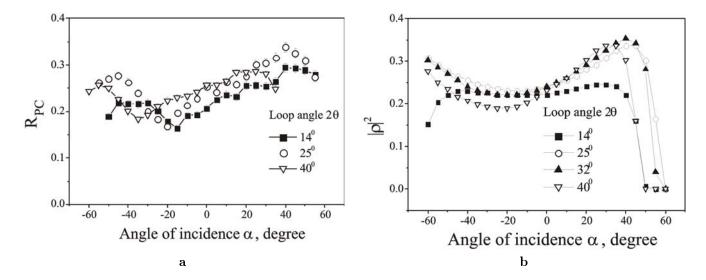


Fig. 3. PC-reflectivity of the ring-loop phase conjugate mirror with  $Sn_2P_2S_6$  crystal at various loop angles  $2\theta$  and the incidence angles  $\alpha$ : a — experiment, b — calculations

mainly on the material parameters of PR crystal and well correlates with the photorefractive build-up time measured by the two-wave mixing.

The results of the measured PC reflectivity in dependence of the incidence angle  $\alpha$  measured at several loop angles  $2\theta$  are presented in Fig. 3, a. It is seen that the maximal reflectivity is observed at the loop angle  $2\theta \approx 25^{\circ}$  and the angle of incidence  $\alpha$  near 45°. Evidently, this maximum is a result of the compromise between decreasing the effective electro-optic (EO) coefficient due to rotation of the grating vector K away from the polar axis and falling the two-wave mixing coefficient, and increasing the sample optical transmission due to decreasing the light reflectivity by the crystal faces at approaching the Brewster angle (about  $70^{\circ}$  in our case). This situation will be analyzed below in details.

The measured PC wave build-up time  $\tau_{PC}$  in dependence on the both  $\theta$  and  $\alpha$  angles is shown in Fig. 4,a. The  $\tau_{PC}$  can be considered as a value proportional to the PR response time  $\tau_{PR}$ . In our case of the dominating diffusion mechanism of the PR effect [16], this can be expressed as

$$\tau^{-1} \approx \tau_{\rm PR}^{-1} = \frac{C(\Lambda)}{\varepsilon \varepsilon_0} (\gamma_D + \gamma_{\rm ph} I_0),$$
(2)

where

$$C(\Lambda) = \frac{1 + (2\pi l_S/\Lambda)^2}{1 + (2\pi L_D/\Lambda)^2}.$$

Here,  $\gamma_{\rm D}$  is the dark conductivity,  $\gamma_{\rm ph}$  is the photoconductivity,  $\Lambda$  is the space-charge grating spacing

 $(K=2\pi/\Lambda)$ ,  $l_S$  is the Debye screening length, and  $L_D$  is the diffusion length. Admitting both  $\gamma_D$  and  $\gamma_{\rm ph}$  to be isotropic values, the variation of the time characteristic is defined by the corresponding dielectric constant  $\varepsilon$  and the grating spacing  $\Lambda$ .

## Numerical Model of the Phase Conjugation

A theory of the ring-loop oscillators developed in [2] under the assumption of negligible optical absorption and depleted pump (i.e. relatively high two-wave mixing gain coefficient  $\Gamma$ ) gives the following relationships:

$$\frac{\left|t_{0}\right|^{2}\operatorname{tgh}\left(k\right)}{s+\sigma\operatorname{tgh}\left(k\right)} = \frac{s-I_{0}\operatorname{tgh}\left(k\right)}{\left(\sigma I_{0}-s^{2}\right)\operatorname{tgh}\left(k\right)+\left(I_{0}-\sigma\right)s},$$
(3)

where

$$\sigma = I_0 \frac{\left|t_0\right|^2 - 1}{\left|t_0\right|^2 + 1}, \quad s = \sqrt{\sigma^2 + (I_0 - \sigma)^2 \left|\rho\right|^2}; \quad k = \frac{s\Gamma l}{4I_0},$$

 $|t_0|^2$  is the loop transmission,  $\Gamma l$  is the production of the two-wave mixing gain  $\Gamma$  and the interaction length (crystal thickness) l, and  $|\rho|^2 = R_{\rm PC}$  is the PC reflectivity.

As was shown in [17], this approximation is valid also for absorbing crystals, when the optical absorption coefficient does not exceed 0.1  $\Gamma$ , that takes place in our case. Note that here we consider only the "transmission" grating (formed by beam pairs 1-2 and 3-4 in Fig. 1,b).

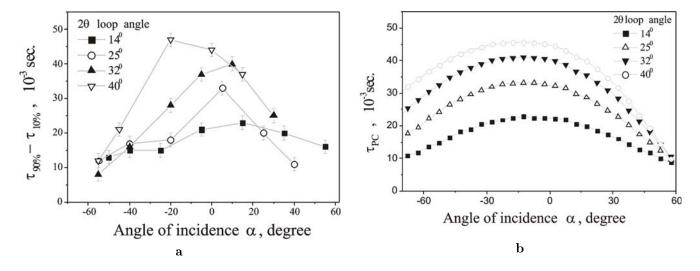


Fig. 4. PC-beam build-up time in the ring-loop phase conjugate mirror with  $Sn_2P_2S_6$  crystal as a function of the incidence angle  $\alpha$  at various loop angles  $2\theta$ : a—experiment, b—calculations

Basing on Eq. (3), we analyzed numerically the PC reflectivity measured above with aim to build up a model of the studied SPPC scheme with the  $\rm Sn_2P_2S_6$  PR sample. To calculate the PC reflectivity in dependence on the sample orientation and position, Eq. (3) should be solved numerically using the data on the two-mixing gain  $\Gamma l$  and refractive index n at different values of the input beam incidence angle  $\alpha$  and the loop angle  $\theta$ . For each  $\alpha$  and  $\theta$  we calculated the modulus and orientation of the space-charge grating vector k. The variation of the effective crystal thickness and the Fresnel reflection were accounted in optical losses using the absorption coefficient 0.5 cm<sup>-1</sup> measured in the studied sample at  $\lambda = 633$  nm. The main values of the refractive indices at this light wavelength are 3.024, 2.930, 3.094 [18].

The most problematic is an analytical expression for the orientation dependence of the gain coefficient Γ. As was shown before [9], the dependence of the gain on the grating spacing is described in the framework of the diffusion mechanism with the presence of two types of charge carriers and the partial electronhole compensation. In this case, holes form the fast component and electrons form the slow one of the photoinduced space-charge grating. We used the following expressions for the two-wave mixing gain:

$$\Gamma = AK \left( \frac{1}{1 + B^{-2}K^2} - \frac{1}{1 + C^{-2}K^2} \right),\tag{4}$$

$$A = r^{\text{eff}} \frac{2\pi n^3 k_{\text{B}} T}{e\lambda},\tag{5}$$

$$B = e \left(\frac{\lambda}{4\pi}\right)^{3} \left(\frac{N_{R1}}{\varepsilon \varepsilon_{0} k_{B} T}\right)^{1/2},$$

$$C = e \left(\frac{\lambda}{4\pi}\right)^{3} \left(\frac{N_{R2}}{\varepsilon \varepsilon_{0} k_{B} T}\right)^{1/2}.$$
(6)

Here, e is the elementary charge,  $k_{\rm B}T$  is the thermal energy,  $\varepsilon\varepsilon_0$  is the dielectric permittivity, and the  $N_{R1}$  and  $N_{R2}$  are the hole and electron effective trap densities taken as the isotropic values and determined by fitting. Also by fitting, we obtained the EO coefficient value for a space-charge field parallel to the OX axis (i.e.  $r_{111}$ ). This number was taken to be 110 pm/V in good correlation with direct measurements [19] and previous estimations [9, 11]. The angle-dependent values are the  $r^{\rm eff}$ , dielectric constant  $\varepsilon$ , the refractive index n, and the PR grating wave vector K. The orientation dependence of the two-wave mixing coefficient  $\Gamma$  was calculated using (4)—(6) with these parameters.

Unfortunately, not all components of the EO tensor of the  $\mathrm{Sn_2P_2S_6}$  are measured to date. Even the estimated relationships between components of the electro-optic tensor [20] are not known so precisely to make it possible to calculate the orientation dependences of  $r^{\mathrm{eff}}$  in the XZ plane. To solve this problem, we measured directly the angular dependence of the two-wave mixing gain  $\Gamma l$  on the Y-cut of the studied sample. These measurements were carried out with turning the  $\mathrm{Sn_2P_2S_6}$  sample when the azimuth of polarization of the interacting beams was rotated simultaneously. The gain factor measured in such a way follows the variation of the effective EO coefficients when the electric vector of the space-charge

grating rotates from X to Z. At the same time, the polarization azimuth of the interacting beams at each point corresponds to one of two main axes of the Fresnel ellipsoid (X', Z'). Note that these main axes in  $\operatorname{Sn_2P_2S_6}$  at room temperature and  $\lambda = 633$  nm are turned by  $45^\circ$  relatively to the X and Z axes in the (010) mirror plane. The angle  $\varphi_0$  on this dependence, that corresponds to the compensation of the EO coefficient and  $\Gamma(\varphi_0) = 0$ , gives us the relationship  $r_{113}/r_{111} = \operatorname{tg} (\varphi_0) \approx 0.06$  used in our calculations. Using this number, we approximated the angular dependence of the effective EO coefficient in the XZ plane (neglecting the  $r_{131}$  and  $r_{133}$  components) as follows:  $r^{\text{eff}} \approx r_{111} \cos \alpha' + r_{113} \sin \alpha'$ , where  $\alpha'$  is the angle between the space-charge grating vector and the OX crystal axis.

The angular dependence of the dielectric constant was calculated accounting the main values of the tensor ( $\varepsilon_{11} = 235$ ,  $\varepsilon_{33} = 65$  [21]), and the angle between the dielectric tensor main axes and the above coordinate system is 14° in the XZ plane [22].

Using the above procedure and values, the calculated PC reflectivity at various  $\alpha$  and  $\theta$  angles is shown in Fig. 3,b. On the whole, a good correlation of the measured and calculated data is observed. The difference in the curve shapes is marked mainly near the angle  $\alpha = -10...-20^{\circ}$  (Fig. 3). The lower measured value of the PC reflectivity can be explained by a partial erasing of the PR grating by the laser beam partially reflected inside the sample into the region of the interaction.

The same dependences of the PC rise time were also calculated according to (2). The curves  $\tau_{PC}$  vs.  $\alpha$  (Fig. 4,b) were obtained at various  $\theta$  and reflect the variations of both the direction and the modulus of the grating spacing wave vector K and are in some correlation with the experimental data shown in Fig. 4,a. They were calculated using the same material parameters as above, and some additional material parameters (dark- and photoconductivities  $\gamma_D$  and  $\gamma_{\rm ph}$ , screening length  $l_S$ , and diffusion length  $L_D$ ) were determined from the direct measurements of the twowave mixing dynamics in the studied sample by the method used before in [9, 11]. The following parameters were obtained:  $\gamma_D = 1.7 \cdot 10^{-9} \ (\Omega \cdot \text{cm})^{-1}, \ \gamma_{\text{ph}} = 359$  $\text{cm}/(\text{W}\cdot\Omega)$ ,  $l_S = 0.24 \,\mu\text{m}$ ,  $L_D = 0.78 \,\mu\text{m}$ . Note that these data correspond to the "fast" components of the PR grating, because the time constant was measured on the initial stage of the PR grating formation, whereas the saturated PC reflectivity is formed after charge compensation.

Comparing the data presented in Figs. 3 and 4, it is seen that the higher PC reflectivity corresponds to the

relatively short rise time. This is the sequence of the rather favorable mutual orientation of the main axes of the material tensors in  $\mathrm{Sn_2P_2S_6}$  which determine the values and anisotropy of  $R_{\mathrm{PC}}$  and  $\tau_{\mathrm{PC}}$ . The PC wave rise time can vary by about four times by adjusting the sample orientation and loop angle. In principle it is possible to get a faster response in the thicker sample cut off at some angle to the conventional one (used above) on the XZ plane, even in spite of the lowering of the effective EO coefficient. Nevertheless, the used crystal geometry is close to the optimal one.

The above-presented numerical model allows us to evaluate the influence of the sample thickness on the maximal PC reflectivity in the used scheme. It was obtained that the sample thickness reduced to 4 mm will leads to the reflectivity decreasing by 5%. Increasing the PC reflectivity by the same value requires the sample thickness above 12 mm.

The approach developed above can be used to optimize the active element of the SPPC mirrors based on  $\rm Sn_2P_2S_6$  crystal with special shapes, with the antireflecting coating, and other factors.

### Conclusion

Thus, we measured the dependence of the PC reflectivity and the conjugate wave rise time in the ring-loop SPPC mirror using  $\mathrm{Sn_2P_2S_6}$  crystal as a PR medium on the geometry of experiment at  $\lambda=633$  nm. It was shown that the dependences of these parameters on the crystal geometry are well described by the theory [2] using the material parameters of the crystal. The developed numerical model can be used upon choosing the optimal crystal cut, sample thickness and other "geometric" parameters for various modifications of  $\mathrm{Sn_2P_2S_6}$  ("brown" samples, doped crystals, etc.), as well as upon using the sample with antireflecting coating in order to achieve the best performance of the reflectivity and (or) the PC wave rise time of the studied scheme.

We dedicate this work to Prof. M.S. Soskin on the occasion of his 75th birthday.

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#### **ПЕТИМИТПО**

ПЕТЛЬОВОГО ГЕНЕРАТОРА, ПОБУДОВАНОГО НА ФОТОРЕФРАКТИВНОМУ КРИСТАЛІ  ${\rm Sn_2P_2S_6}$ 

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Резюме

Досліджено основні параметри петльового фазообертаючого дзеркала в залежності від експериментальних змін геометрії взаємодії. Запропоновано чисельну модель, що описує динамічні характеристики і зміну відбивної здатності фазообертаючого дзеркала. Цю модель можна застосовувати для оптимізації властивостей фоторефрактивних елементів, побудованих на кристалі  $Sn_2P_2S_6$ .