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13. ABSTRACT (Maximum 200 words)

This report results from a contract tasking Dynamic Holography Group, Institute of Physics. National Academy of Sciences as follows: The contractor will investigate getting the largest possible phase conjugate reflectivity in Sn2P2S6 by optimizing the usual parameters (such as pump intensity ratio, signal to pump intensity ratio, angle of incidence for pump and probe waves, polarization of interacting waves, orientation of waves with respect to sample principle axes) and frequency shift, a new parameter of special importance of Sn2P2S6. To achieve this the contractor shall perform the following: (1) The backward wave from four wave mixing will be used to generate the phase conjugate waves. Optimum conditions will be found to accomplish this. (2) Study different self-phase conjugate mirrors based on various coherent optical oscillators Optimize the performance of a double phase conjugating mirror. The study shows that SPS crystal can be efficiently used for conjugation of coherent light beams both in traditional backward-wave four-wave mixing geometry and in self-pumped coherent oscillator geometries. The samples belonging to Type I are suitable for conjugation both in the red (He-Ne laser light) and in near infrared (Nd^{3+}:YAG, 1.06 μm) while samples of Type II can be used for red and shorter infrared (0.8... 0.9μm) region of spectrum. The phase conjugate intensity twice as large as the signal intensity was easily achieved in the red, at $\lambda = 0.63$ μm. It is inevitable that with the same sample and the same refractive index modulation the phase conjugate reflectivity in infrared diminishes as λ^{-2} with the increasing λ. This decrease can be compensated for by increasing the thickness of the samples used, what is essentially technological and not physical problem. With the available samples we got 17 % phase conjugate reflectivity for Nd^{3+}:YAG, 1.06 μm radiation.

During this study the unusual properties of photorefractive ring-loop oscillator with SPS were revealed: the grating self-developing inside the sample appears to be moving grating and the oscillation wave diffracted from this grating is frequency shifted with respect to the pump wave (because of Doppler effect). This behavior is inherent feature of the laser with photorefractive medium possessing two types of movable charge species; the laser finds himself this mode of operation which is very interesting practically as it ensures the enhanced phase conjugate output compared to traditional four-wave mixing geometry.

From our experience (and from our first experimental attempts undertaken in other laboratories) we can suggest that SPS can be profitable for the wavelength range covered by Ti : Sapphire laser, including 0.8... 0.9 μm where different semiconductor diode lasers are operating.

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Optical Phase Conjugation

with Photorefractive Sn$_2$P$_2$S$_6$

FINAL REPORT

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CONTENT

INTRODUCTION

Nonlinear Four-Wave Mixing in photorefractive materials allows for generation of phase-conjugate light waves at relatively low (down to microwatt) intensity levels (see, e.g., [1]). With the crystals known at present the best performance of Phase Conjugate Mirrors (PCM) was achieved in the visible (green) region of spectrum [2]. At the same time the phase conjugation in Near Infrared would be of great interest because of possible applications with well developed diode-pumped Nd^{3+}:YAG lasers. Laser-beam clean-up, image deblurring, optical correlation are only few examples of what can be done.

Several laboratories all over the world are working now in this field trying to solve the main problem which is the problem of suitable material. The known photorefractive semiconductors ensure quite fast response with poor gain factor [3] while recently developed rhodium-doped barium titanate [4,5] provides rather high gain with considerably longer response times (hundreds of seconds).

The purpose of this project is to use different wave-mixing processes in new photorefractive material, tin hypothiodiphosphate, $Sn_2P_2S_6$, [6,7] to generate the phase conjugate waves in the red and near infrared region of spectrum.

Our previous study of two-beam coupling in $Sn_2P_2S_6$, undertaken together with the group of Dr. George Brost from former Rome Laboratory [8,9] showed that this material combines relatively fast response (milliseconds range) with sufficiently large gain factor (7... 8 cm^{-1}). At the same time the presence of two out-of-phase photorefractive gratings in $Sn_2P_2S_6$ does not allow for trivial extension of the known techniques of phase conjugation to this material.

The main goal of the project is to get the largest possible phase conjugate reflectivity in $Sn_2P_2S_6$ by optimizing the usual parameters (as pump intensity ratio, signal to pump intensity ratio, angle of incidence for pump and probe waves, polarization of interacting waves, orientation of waves with respect to sample principal axes) and frequency shift, a new parameter of special importance for $Sn_2P_2S_6$.

MATERIAL CONSIDERATION AND EXPERIMENTAL PROCEDURE

Photorefractive $Sn_2P_2S_6$

Tin Hypothiodiphosphate ($Sn_2P_2S_6$, SPS) belongs at ambient temperature to the monoclinic symmetry, possessing unique mirror symmetry plane (normal to Y axis) [10]. Its axis of spontaneous polarization P_s makes an angle about $10°$ to the crystal X-axis in symmetry plane. The largest electrooptic constant (from the evaluated at present) is $r_{111} \approx 75$ pm/V ; therefore we use in what follows the interaction geometry with r_{111} involved in readout of photorefractive grating. In other words, Z-cut samples are used and interacting waves with polarization nearly parallel to X-axis are sent to the sample to form the transmission gratings with grating vector parallel to X-axis.

To sensitize the samples for the recording in Near Infra Red we use pre-illumination with light. A 100W halogen lamp light is guided to the sample with flexible bunch of light waveguides; the exposure time is about 5 minutes. From our own experience [9] we know that illumination of this kind is sufficient to keep the sample in sensitized state as minimum during total working day.

As grown crystals quite often possess a high degree of polarization, i.e., the amount of domains with certain orientation of spontaneous polarization is much higher than the number of domains with opposite ($180°$) orientation of spontaneous polarization. To ensure the correct comparison of data for different sample we developed the poling technique which was used for most our samples. First the sample is slowly heated ($20°C$ per hour) up to $100°C$ (phase transition temperature is about $60°C$). Next the sample is exposed to white light during 3 hours at $100°C$. Heating and illumination are increasing the crystal conductivity and are supposed to smooth out any large scale nonuniform space charge. Then, the light is switched off and the voltage (about 1500 V/cm) is applied to the sample which is slowly cooled down to the ambient temperature ($15°C$ per hour). The silver paint contacts were deposited to [100] faces of the sample to apply the voltage. Finally at room temperature the sample with no applied voltage is one more time exposed to white light during three hours. It is not clear whether the sample becomes perfectly single-domain after this treatment but the degree of its poling is always largely improved what can be seen from the enhancement of the beam coupling.

Sample characterization

As a first step the characterization of the available SPS sample have been performed. The samples used [11] were nominally undoped but cut from the ingots grown in slightly different conditions, sometime with different after-growth treatment and different poling technique. All these factors influence photorefractive properties rather strongly; therefore the pre-selection should be done for finding the sample convenient for Phase Conjugate wave generation.

A standard technique of two-beam coupling in the course of transient grating recording was used to extract the data on the Debye screening length (effective trap density), diffusion length, characteristic grating decay time, and to evaluate the phenomenological parameter - gain factor

$$\Gamma_e = \frac{1}{\ell} \ln \frac{I_S(\ell) I_P(0)}{I_S(0) I_P(\ell)}, \qquad (1)$$

where ℓ is the sample thickness, I_s and I_p are the intensities of the signal wave and the pump wave, respectively. The gain factor is directly proportional to the ultimate refractive index change and can be used to estimate the efficiency of the phase conjugation in different Four-Wave-Mixing geometries.

The schematic drawing of the experimental set-up for sample characterization is shown in the Fig.1. As a source of coherent light we used either cw He-Ne laser ($\lambda = 0.63$ μm, 40 mW output power, TEM_{00}) or cw diode-pumped Nd^{3+}:YAG laser ($\lambda = 1.06$ μm, 500 mW output power, single frequency, TEM_{00}). With the semitransparent mirror M2 two recording beams were formed and sent to the sample at an angle 2θ. The fringe pattern with fringe spacing Λ

$$\Lambda = \lambda \ / \ 2 \sin\theta, \qquad (2)$$

appears in the sample, giving rise to the space charge redistribution and formation of the photorefractive grating. The phase retarder ($\lambda/2$-plate) and polarizer in front of the mirror M2 are used to control the recording beam intensity. The shutter in one arm of the interferometer is used to stop the pump wave and analyze the light induced erasure of the photorefractive grating.

The intensity of the weak signal wave is monitored during the recording-erasure cycle with the help of photodiode; the signal is fed to the PC via analog-digital converter and stored in PC memory. The whole procedure (shutter control, intensity acquisition, data storage) is fully automated.

Figure 2 shows the typical temporal evolution of the weak signal beam intensity ($\lambda = 1.06$ μm) when the virgin sample (all previously recorded gratings are erased by uniform illumination) is exposed to two recording waves, pump and signal. The data on SPS sample #K3 are presented in Fig.2a with grating spacing $\Lambda = 0.9$μm, $I_p = 20$ W/cm^2, $I_s = 0.2$ W/cm^2, while Fig.2b shows the data for sample #K2 with grating spacing $\Lambda = 3.6$ μm, $I_p = 20$ W/cm^2, $I_s = 0.2$ W/cm^2 .

All samples tested had one of two kinds of response [12], one with the pronounced transient amplification (Fig.2a) and the other with smooth build-up of a gain until the steady state is reached (Fig.2b). The appearance of transient peak in two-beam coupling gain for samples of Type I is attributed to formation of the out-of-phase photorefractive grating by thermally excited carriers [7,9]. This grating compensates partially for the grating recorded via light-induced charge transport and therefore inhibits the steady-state gain. For the samples belonging to Type II the thermally excited charge motion is inefficient, either it does not exist at all or is too slow to detect it within reasonable measuring time. We call the steady-state gain factor which is due to the "fast" grating Γ_e and the steady-state gain factor which is due to the "slow" thermally excited grating Γ_h; in the same manner the relaxation time of the "fast' grating is called τ_e and that of slow grating τ_h. (In fact, we do not know at present which one is formed by the electrons and which one by holes, so the letters used for subscripts here are meaningless).

Figures 3a,b represent the fringe spacing dependences of the measured gain factor for the sample #K3 at $\lambda = 0.63$ μm and $\lambda = 1.06$ μm, respectively (filled squares). In the experiment we are working always with the unexpanded light waves; for large angles between the recording waves this results in incomplete overlap of two beams inside the crystal and to limitation of the interaction length which becomes smaller than the sample thickness (Fig.4). To account for this factor when evaluating the gain factor we put the corrected values of interaction length in following way

$$\Gamma = \Gamma(\text{measured}) \ / \ \zeta, \qquad (3)$$

$$\zeta = 1 - (\ell \ /d) \tan \theta', \qquad (4)$$

where d is the Gaussian beam waist, θ' is the half-angle between the recording waves inside the sample, and ℓ is the sample thickness.

The open squares in Fig.3a,b are the measured data corrected in accordance with Eqs.3,4. Note, that the procedure described is changing the position of the maximum of the gain factor in fringe spacing scale; therefore the Debye screening length evaluated from these data also changes.

Table 1 summarizes the results of characterization for two samples, representing two types of SPS crystals [12].

(Table 1)

sample #	Dimensions (a × b × c), mm^3	Wavelength, μm	Gain factor, cm^{-1}	Debye screening length, μm
K2	5 × 5 × 2	0.63	11	1
		1.06	0.9	6.4
K3	9 × 4.5 × 9	0.63	6	1.7
		1.06	3.2	1.3

The comparison of sensitivity for crystal of two types showed that only Type I samples have a relatively good response at λ = 1.06 μm, with the gain factor (corrected for interaction length) reaching Γ ≈ 8 cm^{-1} (Fig.3b). Therefore we were working in present study mainly with samples belonging to Type I.

Surprising feature of SPS as compared to other photorefractive crystals is rather unexpected intensity dependence of the gain factor. In addition to the well known intensity dependence which is due to the relatively high dark conductivity the slow increase of Γ_e is observed even in the intensity range where photoconductivity largely exceeds the dark conductivity (see the intensity range larger than 2 W/cm^2 in Fig.5a,b).

The behavior is similar for λ = 0.63 μm, Λ = 1.44 μm (Fig.5a) and for λ = 1.06 μm, Λ = 1.2 μm (Fig.5b). In our measurements shown in this Report we always try to use the largest possible intensity of the unexpanded laser beams to get larger Γ_e and higher phase conjugate reflectivity. It should be taken into account, however, that we were never able to saturate Γ_e with the power of our lasers, so the higher values of Γ_e than that reported are possible, in principle.

Experimental set-up for phase conjugation

Two geometries were used: a classical backward-wave four-wave mixing geometry (Fig.6) and a self-pumped optical conjugator based on the ring-loop coherent optical oscillator (Fig.7). The first geometry differs from that of two-beam coupling (Fig.1) by the presence of an additional pump wave, counterpropagating to the recording pump wave. This auxiliary light beam 2 reads out the photorefractive grating recorded by a pair of copropagating (signal 3 and pump 1) waves and the wave 4 diffracted from the grating is just the phase conjugate replica of the signal wave. The temporal frequencies of all tree beams which impinge upon the sample (1,2, and 3) are identical, therefore the frequency of the generated phase conjugate wave (4) is expected to be also the same (to meet the conservation law for degenerate four-wave mixing).

The output beam of the laser is first splitted in two parts by the semitransparent mirror M2. More weak beam is sent to the sample with mirrors M3 and M6 as a signal wave 3. Both mirrors M3 and M6 are partially transparent to detect the generated phase conjugate beam, counterpropagating to the signal one. The intensity of the phase conjugate beam is measured by a photodiode (with amplifier) and sent to PC. The diaphragm ensures spatial filtering of conjugate beam only to be sent to detector.

The beam transmitted through the mirror M2 is directed to the polarizer beam splitter after passing the phase retarder (λ/2-plate). With this arrangement it is easy to control the intensity ratio of two splitted beams. The mirrors M4 and M5 were sending two waves to the sample, forming two counterpropagating pump waves 1 and 2. An additional phase retarder (λ/2-plate) was placed in one arm of this interferometer to adjust the polarization of both beams to be identical and parallel to the sample X-axis. The shutters Sh2 and Sh1 allowed for cutting the signal beam or all incident beams, respectively.

The oscillator geometry (Fig.7) involves only one light beam; this beam after passing through the sample is redirected to the same area in the sample by two cavity mirrors, M3 and M5. The semitransparent mirror M4 is used to reflect the backpropagating phase conjugate wave in direction of photodiode. An additional mirror M2 is used to form a reference wave to analyze possible frequency shift of the oscillation wave; this reference wave can be cut by a shutter placed in front of mirror M2.

As for any other oscillator for this one the threshold conditions of selfoscillation should be met, both the amplitude condition (gain overcome losses) and the phase condition (phase difference between two copropagating waves, pump and oscillation, should remain the same after each round trip in the cavity). These threshold conditions impose certain restrictions on the sample thickness, angle between the waves copropagating inside the sample and frequency detuning of the oscillation wave (as two of four interacting waves selfdevelop in this geometry from the noise they may have the frequency close but not exactly the same as frequency of the pump).

EXPERIMENTAL RESULTS

Phase conjugation with backward wave four-wave mixing geometry

The presence of two out-of-phase gratings for samples of Type I was expected to affect also the dynamics of the phase conjugation in backward-wave four-wave mixing. The temporal evolution of the intensity of the phase conjugate wave presented in Fig.8a clearly shows a pronounced peak immediately after the beginning of exposure and gradual decrease of intensity until the steady state is reached (The grating with grating spacing $\Lambda = 0.9$ μm is recorded and the pump ratio is $r = 0.18$).

We were aware whether this behavior can be attributed to formation of out-of-phase grating only, or the important light-induced scattering (beam fanning) can also be the reason of the decrease of steady state intensity of the phase conjugate wave. To check it we compare the dynamics of phase conjugate intensity in two experiments with different initial conditions: when all three interacting wave start to illuminate the virgin crystal and when crystal is first exposed to two pump wave (in order to stabilize the beam fanning from pump waves) and then the signal wave is sent to the crystal. From this comparison we conclude that more than 80% of the decrease of the phase conjugate wave intensity can be attributed to the formation of out-of-phase grating. In what follows in this section all the measurements are performed with pre-illumination of the sample with two pump waves. Fig.8b represents the dynamics of phase conjugate wave calculated from the set of material equations; it will be discussed in the next section.

From the data similar to that of Fig.8a the peak value of phase conjugate intensity was used to calculate the phase conjugate reflectivity (normalized to the intensity of the incident signal wave), R_{pc}. Further on, we plot the dependence of the phase conjugate reflectivity R_{pc} as a function of pump intensity ratio $r = I_{p1} / I_{p2}$ at $\Lambda = 1.6$ μm (Fig.9). The largest reflectivity is measured for the unequal intensities of the pump waves $r \neq 1$ as one can expect for the four-wave mixing in a medium with $\pi/2$-shifted index gratings. The angle 2θ between the signal and copropagating pump wave is chosen to be $2\theta = 36°$ to ensure the largest gain factor (as measured from two-beam coupling experiments). The direct measurements of fringe spacing dependence of phase conjugate reflectivity confirmed the correctness of this choice (Fig.10). The effective coupling strength reduced for sample losses is $\Gamma \ell = 2.4$.

The phase conjugate reflectivity becomes larger for smaller intensities of the signal wave (Fig.11), so we tried to chose the signal-to-pump intensity ratio corresponding to the plateau in dependence shown in this figure.

With all parameters optimized in the described way we got the amplified phase conjugate output (intensity of the reflected phase conjugate wave was larger than that of the incident signal wave) with $R_{pc} = 200\%$. The frequency of the phase conjugate wave in the described experiments was exactly the same as frequency of both pump waves and signal wave (strict degeneracy).

Phase conjugation with ring-loop oscillator

The dynamics of the phase conjugate wave intensity in this case (Fig.12a,b) is typical for photorefractive oscillators. For relatively long time the phase conjugate wave is hardly distinguishable as compared to the components of light-induced scattering propagating in the same direction ($t < t_{on}$ in Fig.12), but after a certain well defined temporal threshold the nonlinear growth of the phase conjugate wave intensity occurs until the saturation is established. Note two features distinguishing this dynamics from that for backward-wave four-wave mixing (Fig.8a) described in previous section: (i) there is no transient peak (steady-state intensity is reached smoothly) and (ii) intensity oscillations appear, either damped (well pronounced at the beginning of exposure) like in Fig.12a or steady-state like in Fig.12b . The curves of Fig.12a,b are recorded for different grating spacing ($\Lambda = 1.6$ μm and $\Lambda = 3.2$ μm, respectively) but keeping the same incident intensity $I = 3$ W/cm^2 . This explains the difference in phase conjugate reflectivity and modulation frequency.

To get the largest phase conjugate reflectivity we optimize for the angle of the ring-loop oscillator $2\theta = 36°$ as it has been described in the previous section and use high quality dielectric cavity mirrors with minimum transmission losses.

To evaluate the possible frequency shift of the phase conjugate wave with respect to the incident wave from the laser the beat frequency mark is recorded when mixing the generated wave with copropagating reference wave (local oscillator), see Fig.7. Figure 13 shows a typical beat frequency mark proving unambiguously the nondegenerate nature of background four-wave mixing process. The detected frequency shift ω was a function of the incident light intensity (usual for photorefractives where relaxation time is inversely proportional to the light intensity) and of spatial frequency of the photorefractive grating selfdeveloping in the sample (Fig.14).

In same cases (very rarely, in fact) the oscillation dynamics had much more pronounced intensity modulation as shown in Fig.12b. The frequency of modulation in that cases was twice as large as beat frequency in case of smooth dynamics. This points to simultaneous excitation of two oscillation waves with different frequencies. We should say, however, that this regime was neither stable nor reproducible and the conditions to get it are not yet well defined.

The ultimate phase conjugate reflectivity reached in this experiment is R_{pc} = 17 % (Fig.15). It looks modest as compared to R_{pc} = 200% for backward-wave four wave mixing but in fact the efficiency of conjugation is much higher for oscillator because the powerful pump waves are not used in this case.

Discussion and comparison with calculation

The results of the experiments on backward-wave four-wave mixing can be compared with calculations made within the undepleated pump approximation [13]. For purely nonlocal response ($\pi/2$-shifted index grating) the amplitude phase conjugate reflectivity (ρ) should be

$$\rho = -\frac{\sinh\left(\dfrac{\dfrac{A\Lambda}{\Lambda^2 + \ell^2_{Sn}}\ell - \alpha'\ell}{4}\right)}{\cosh\left(\dfrac{\dfrac{A\Lambda}{\Lambda^2 + \ell^2_{Sn}}\ell - \alpha'\ell}{4} + \dfrac{1}{2}\ln r\right)} , \qquad (5)$$

where

$$A = -\frac{4\pi^2 n^3 r_{eff}}{\lambda\cos\theta'} \times \frac{k_B T}{e} , \qquad (6)$$

α is the sample absorption constant, θ' is the angle between the writing waves inside the sample, r is the effective electrooptic constant, n is the refractive index (r_{111} in our case), k_B is Boltzmann constant, T is an absolute temperature, e is the electron charge and ℓ_{sn} is Debye screening length for carriers forming the "fast" grating.

For optimized pump intensity ratio, $\ln(r) = (\Gamma\ell - \alpha\ell)/2$, Eq.5 simplifies

$$\rho = -\sinh\left(\frac{\dfrac{A\Lambda}{\Lambda^2 + \ell^2_{Sn}}\ell - \alpha'\ell}{4}\right) . \qquad (7)$$

The solid line in Fig.9 represents the calculated from Eq.5 dependence $R_{pc} \equiv |\rho|^2 = f(r)$ for the effective $\Gamma\ell$ = 4.1 reduced to account for all losses of the sample. The qualitative agreement with the experiment (squares) is rather good; nevertheless the increasing systematic deviation for increasing pump ratio r is obvious. This small discrepancy may result from the intensity dependence of the gain factor. The experimental dependence is measured when keeping the total intensity of two pump waves constant (the polarization beam splitter redistribute the intensities of two light beams when the angle of polarization is changed but total intensity of two beams is preserved). The intensity of two recording waves in this configuration is not constant but diminishes with the increasing pump ratio r. This is, in our opinion, the main reason of the discrepancy, increasing with the increasing pump ratio. When appropriate correction for changing gain factor is introduced the experimental data (filled dots) become much closer to calculated (solid curve) but small quantitative difference still exists. The reason of remaining discrepancy can come from Gaussian profile of the intensity distribution of interacting waves, from incomplete overlap of interacting beams, etc.

It should be noted that the obtained agreement with the calculation when no adjustable parameter is used can be considered as rather good. Extrapolating the results obtained one can predict the phase conjugate reflectivity 1000 % for $\Gamma\ell$ = 7.5 and 10

000 % for $\Gamma \ell = 12$. It is not easy, however, to reach these values for unexpanded laser beams because the interaction length ℓ becomes smaller than the sample thickness for too thick samples.

There is no theory for nondegenerate oscillation in the ring-loop cavity to compare with for our experimental results. The purely degenerate oscillation is well described in [13]. The small deviations from strict degeneracy were considered in several papers (see, e.g. [14,15]) but they were always the consequence of nonreciprocity inside the laser cavity itself, either introduced artificially with Faraday cell [14] or by tilting the cavity mirror [15].

We performed an analysis of oscillation conditions for particular case of a medium with two types of movable charge species (one photoexcited and other thermally excited) based on our previous calculations of the gain spectra for nearly degenerate two-beam coupling in such a medium [16, 17]. The theory was developed solving only the material equations, i.e., neglecting possible beam-coupling effects. This approximation, nevertheless suits perfectly well for the description of the threshold of coherent oscillation, when the intensity of oscillation wave is virtually zero and only starts to increase.

The manuscript presenting the results of calculation is now in preparation [18]. We squeeze the final results of this study to describe the behavior of optical oscillation in ring-loop cavity. To excite the oscillation in any kind of oscillator one need to meet the amplitude and phase condition of oscillation, or, in other words, to ensure the gain with is larger than all losses of oscillator and ensure large enough positive feedback.

Referring to Fig.7a we can formulate the phase condition of oscillation in a following way: Let us suppose that oscillation already occurs. Then the incident wave 2 and the diffracted from the grating wave 3 are forming the fringe pattern inside the photorefractive sample. Let the phase difference between these two waves (which defines the spatial position of fringes) is $(\varphi_3 - \varphi_2)$. The wave 3 being reflected from mirrors M1 and M2 returns back to the sample face as wave 1; the same for wave 2 which comes back to the sample face as wave 4. The phase difference of these two waves is $(\varphi_1 - \varphi_4)$. The condition of positive feedback is that the fringes from waves 2 and 3 coincide with the fringes from waves 4 and 2 (or are mismatched only to small fraction of fringe spacing) :

$$\varphi_3 - \varphi_2 \approx \varphi_1 - \varphi_4 . \qquad (8)$$

Taking into account that wave 4 is in fact the pump wave 2 after propagation of cavity with cavity length L we get $\varphi_4 = \varphi_2 + k_p L$. In similar way $\varphi_1 = \varphi_3 + k_{os} L + \varphi_{NL}$, with $k_p = 2\pi /\lambda_p$ and $k_{os} = 2\pi /\lambda_{os}$ standing for the wavenumbers of pump and oscillation waves, respectively. The difference is in possible appearance of an additional contribution φ_{NL} to the phase of oscillation wave which is due to diffraction from unshifted grating (see, e.g., [19]) which appears usually if frequency of oscillation deviates from that of the pump and photorefractive grating is moving inside the sample. By substituting φ_1 and φ_4 in Eq.8 we get the phase condition in a form

$$k_{os} L - k_p L + \varphi_{NL} \approx 0, \qquad (9)$$

Usually for reasonable cavity lengths 10..... 100 cm the difference $(k_{os} L - k_p L)$ which is due to the slight frequency deviation of the oscillation wave with respect to the pump wave $\lambda_{os} \neq \lambda_p$ is negligibly small, $\approx (2\pi L \Delta\lambda/\lambda^2) \approx 10^{-8}$. This is why the remaining nonlinear phase shift φ_{NL} can be compensated for either by an additional phase shift (if Faraday rotator is put inside the loop [14]) or by excitation of oscillation modes with higher indices (and therefore different wavenumbers) [14, 15]. We do not observe the transverse modes with higher indices in our experiment and no unreciprocity is introduced inside the cavity. So we need to find the natural way to reduce to zero (or to minimize) the nonlinear phase shift φ_{NL} for diffraction from moving grating.

For photorefractive crystals with diffusion mediated charge transport the phase condition of oscillation was fulfilled automatically: there is no nonlinear phase shift for the amplified beam diffracted from the $\pi/2$-shifted index grating (see, e.g., [19]). The amplitude condition of oscillation resulted in a requirement imposed to minimum coupling strength to get the oscillation [13]

$$\Gamma \ell \geq (\Gamma \ell)_{th} = -2 [(M + 1)/(M - 1)] \ln [(M + 1)/2M] , \qquad (10)$$

where M is the efficient reflectivity of cavity mirrors including all kinds of cavity losses.

For nondegenerate four-wave mixing the amplitude condition of oscillation will be the same as given by Eq.10 with $\Gamma \ell$ standing for the real part of complex coupling strength while the phase condition will be in minimizing (to zero, if possible, or to the value much smaller than π) the nonlinear phase shift of the oscillation wave developing because of the diffraction from the moving photorefractive grating.

From our calculations we got the following expressions for the real Re{γ} (defines gain factor, Re{2γ} = Γ) and the imaginary Im{γ} (defines phase shift, Im{γ ℓ } = φ_{NL}) parts of complex coupling constant in photorefractive crystal like SPS with two types of movable charge carriers

$$\text{Re}\{2\gamma\} = \frac{1}{1+\omega^2\tau_e^2}\left(\Gamma_e - \frac{\Gamma_h}{1+\omega^2\tau_h^2}\right), \tag{11}$$

$$\text{Im}\{\gamma\} = \frac{1}{1+\omega^2\tau_e^2}\left(\omega\tau_e\Gamma_e - \frac{\omega\tau_h\Gamma_h}{1+\omega^2\tau_h^2}\right). \tag{12}$$

These formulae are obtained from more general equations within approximation of all transport length much shorter than the fringe spacing and for strongly different characteristic relaxation times for gratings formed by carriers of different sign.

Figures 16a,b represent the whole spectra of Re{2γ} and Im{γ} while Fig.17a,b show the detailed spectra near ω = 0 within the range of detuning frequency where oscillation is possible. One can see that these spectra are interrelated in similar way as the real and imaginary parts of the dielectric constant are related to each other via Krammers Kronig law. The particular consequence of this interrelation is in exact coincidence of the maximum in gain spectrum with zeroth value of imaginary part of coupling constant which defines a nonlinear phase shift. This result is especially important, it says that both the amplitude condition and the phase condition of oscillation are fulfilled simultaneously exactly for the same detuning frequency

$$\omega_{os} = \sqrt{\frac{\Gamma_h\tau_h - \Gamma_e\tau_e}{\Gamma_e\tau_h^2\tau_e}} \approx \sqrt{\frac{\Gamma_h}{\Gamma_e\tau_h\tau_e}}. \tag{13}$$

From the presented spectra of Re{2γ} and Im{γ} one can see that two possible frequency detunings (for +ω and for -ω are indistinguishable: the gain factors are exactly the same for both frequencies and the nonlinear phase which is due to diffraction is zero, for grating moving to the left as well as moving to the right. This suggests the conclusion that two oscillation waves should appear simultaneously, one with positive and the other with negative frequency detuning. This should result in high contrast intensity modulation of output oscillation, with doubled frequency 2ω. In practice the simultaneous oscillation of two modes with symmetric frequencies is rather exceptional. Most often the oscillation occurs only with a single shifted frequency and oscillation dynamics is rather smooth as it is shown in Fig.11a. The reason of this behavior may be related to the stability of multimode and singlemode operation of the considered oscillator which falls beyond the frame of our study. We may only comment that in usual solid-state lasers with the ring cavities the only one of two counterpropagating waves oscillates at any given moment but it can flip into the counterpropagating wave with certain time. One possible reason of the removal of degeneracy may be related to the presence of the built-in inherent static electric field in SPS, which makes the gain spectrum asymmetric.

The abovementioned calculation of temporal development of Δn allowed also for calculation of the dynamics of phase conjugate reflectivity in backward-wave four-wave mixing experiments (see Fig.8a,b). Taking

$$\Gamma = \Gamma_e - \Gamma_h\left[1 - \exp(-\frac{t}{\tau_h})\right], \tag{14}$$

and combining this equation with that for phase conjugate reflectivity (Eq.5) one can calculate the temporal evolution of the phase conjugate wave from

$$\rho = -\frac{\sinh\left(\dfrac{\Gamma_e\ell - \Gamma_h\ell(1 - \exp(-t/\tau_h)) - \alpha'\ell}{4}\right)}{\cosh\left(\dfrac{\Gamma\ell - \alpha'\ell}{4} + \dfrac{1}{2}\ln r\right)}. \tag{15}$$

The following parameters taken from independent experiments were used to plot Fig.8b: $\Gamma_e\ell$ = 3.6, $\Gamma_h\ell$ = 2.8, τ_h = 42 s, Λ = 0.9 μm, and r = 0.18.

PHASE CONJUGATION IN NEAR INFRARED

The experiments similar to those described in previous sections were performed also in IR region of spectrum with diode-pumped Nd^{3+}:YAG laser radiation at 1.06 μm.

Figure 18 shows the results for backward-wave four-wave mixing arrangement. The experimental dependence is presented for phase-conjugate reflectivity on pump intensity ratio (dots) and dependence calculated from Eq. is plotted as solid line, with the gain factor $\Gamma = 2.6$ cm^{-1} evaluated in two-beam coupling experiment. Because of larger wavelength of IR light as compared to He-Ne laser wavelength, the phase modulation ($\pi\Delta n \ell /\lambda$) with the same nonlinear change of the refractive index Δn will be smaller in infrared. As the phase conjugate reflectivity is roughly proportional to square of phase modulation $R_{pc} \propto (\pi\Delta n \ell /\lambda)^2$ nearly four-time decrease of phase conjugate reflectivity is expected in IR as compared to visible region of spectrum. This explains relatively small peak value of calculated dependence (solid line in Fig.17). In fact the measured value of R_{pc} is two times smaller (dots in Fig.17). Thus, the discrepancy between the theory and experiment is more pronounced in infrared but still the phase conjugate reflectivity more than 15 % can be achieved.

We were able to get the coherent oscillation of SPS in ring-loop cavity in infrared region of spectrum, too. This result seems astonishing at first glance because in usual four-wave mixing geometry the phase conjugate reflectivity drops nearly to one order of magnitude when the wavelength is increased from $\lambda = 0.63$ μm to 1.06 μm. It is necessary to take into account, however, that the gain factor itself is proportional only to λ^{-1} (and not to λ^{-2} as R_{pc} does). Therefore, if in visible region of spectrum the coupling constant of the sample is two times bigger than its threshold value, the same sample will oscillate also at $\lambda = 1.06$ μm. The direct measurements of the coupling strength supported the previous statement: even reduced for sample losses the coupling strength was larger than the threshold value for the ring-loop oscillator, $\Gamma \ell = 2.3 > 2.0$. At the same time with this coupling strength there was no hope to get oscillation in Double Phase Conjugate geometry, where the threshold coupling strength $(\Gamma \ell)_{th} \geq 4$ is required.

The dynamics of oscillation is shown in Fig.19. It consists of nearly periodic spikes indicating, probably the competition of two or several cavity modes. The ultimate peak phase conjugate reflectivity with ring-loop oscillator was about 0.01. The fringe spacing dependence of the phase conjugate reflectivity is shown in Fig.20.

CONCLUSIONS

Our study shows that SPS crystal can be efficiently used for conjugation of coherent light beams both in traditional backward-wave four-wave mixing geometry and in self-pumped coherent oscillator geometries. The samples belonging to Type I are suitable for conjugation both in the red (He-Ne laser light) and in near infrared (Nd^{3+}:YAG, 1.06 μm) while samples of Type II can be used for red and shorter infrared (0.8... 0.9μm) region of spectrum. The phase conjugate intensity twice as large as the signal intensity was easily achieved in the red, at $\lambda = 0.63$ μm. It is inevitable that with the same sample and the same refractive index modulation the phase conjugate reflectivity in infrared diminishes as λ^{-2} with the increasing λ. This decrease can be compensated for by increasing the thickness of the samples used, what is essentially technological and not physical problem. With the available samples we got 17 % phase conjugate reflectivity for Nd^{3+}:YAG, 1.06 μm radiation.

During this study the unusual properties of photorefractive ring-loop oscillator with SPS were revealed: the grating selfdeveloping inside the sample appears to be moving grating and the oscillation wave diffracted from this grating is frequency shifted with respect to the pump wave (because of Doppler effect). This behavior is inherent feature of the laser with photorefractive medium possessing two types of movable charge species; the laser finds himself this mode of operation which is very interesting practically as it ensures the enhanced phase conjugate output compared to traditional four-wave mixing geometry.

From our experience (and from our first experimental attempts undertaken in other laboratories) we can suggest that SPS can be profitable for the wavelength range covered by Ti : Sapphire laser, including 0.8... 0.9 μm where different semiconductor diode lasers are operating.
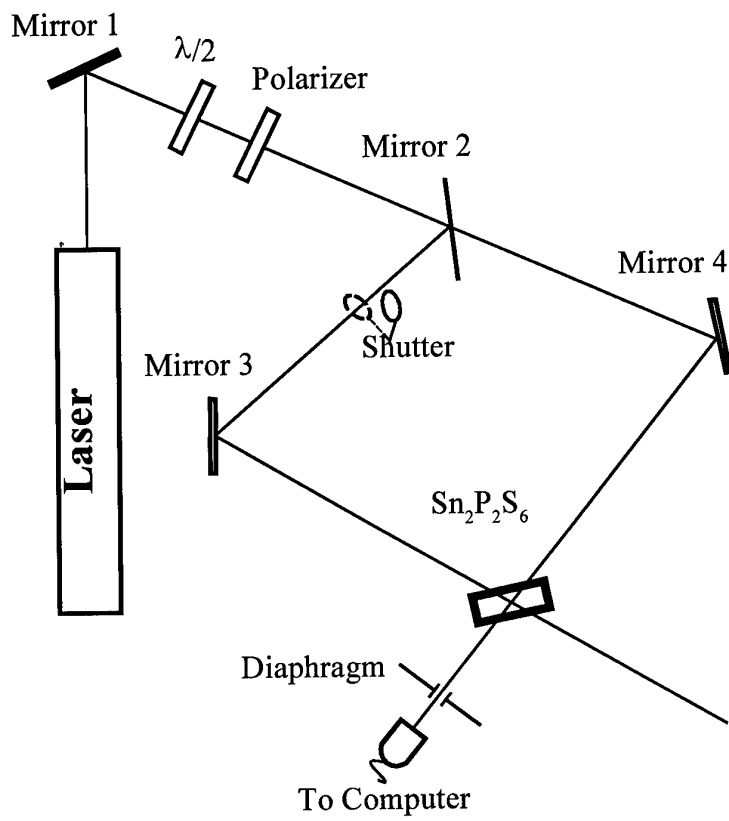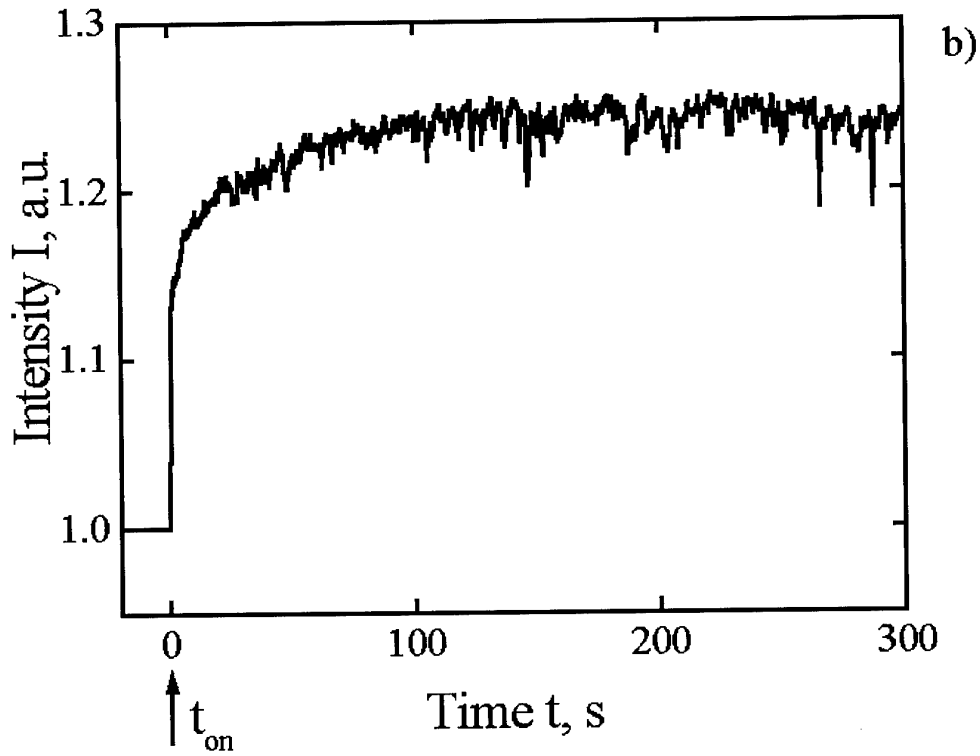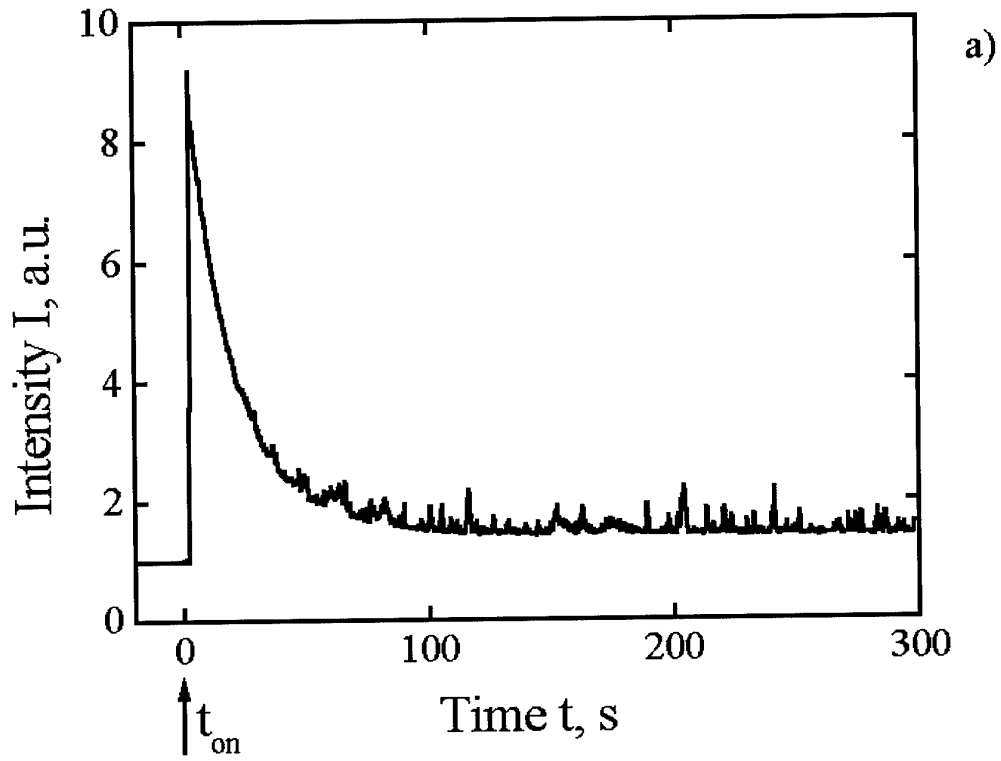
Mirror 1
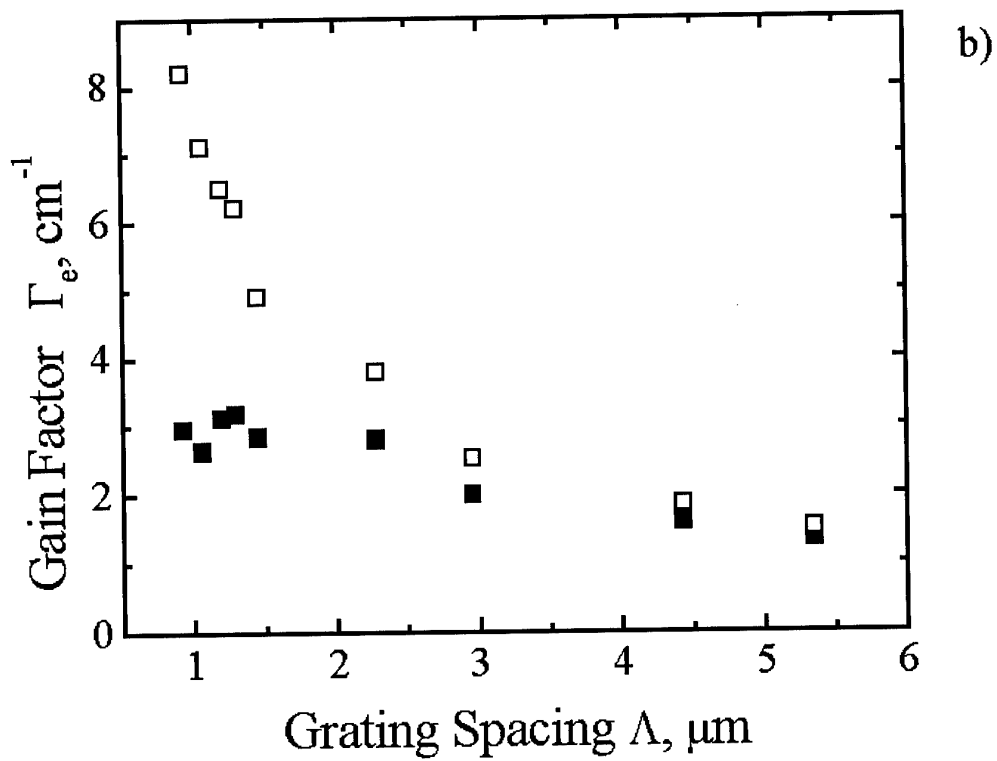
λ/2

Polarizer

Mirror 2

Mirror 4

Shutter

Mirror 3

Laser

$Sn_2P_2S_6$

Diaphragm

To Computer

Fig. 1

Fig. 2

a)

b)

Fig. 4

a)

b)

Mirror 1

Shutter 1

Mirror 2

Mirror 3

Polarizer

$\lambda/2$

Polarizer Cube

Shutter 2

$\lambda/2$

He-Ne Laser

Mirror 4

Mirror 5

$Sn_2P_2S_6$

1

2

3

Diaphragm

4

Mirror 6

To Computer

I_4

I_3

I_2

I_1

Fig. 6

a)

I_4

I_3

I_2

I_1

b)

Mirror 1

Mirror 2

Diaphragm 2

Shutter

Mirror 3

Mirror 4

Polarizer

Diaph. 1

1

$Sn_2P_2S_6$

3

2

Lens

Photodiod

Mirror 5

To Computer

Laser

Fig. 7

a)

b)

Fig. 10

Fig. 15

Fig. 16

Fig. 18

Fig. 19

Fig. 20

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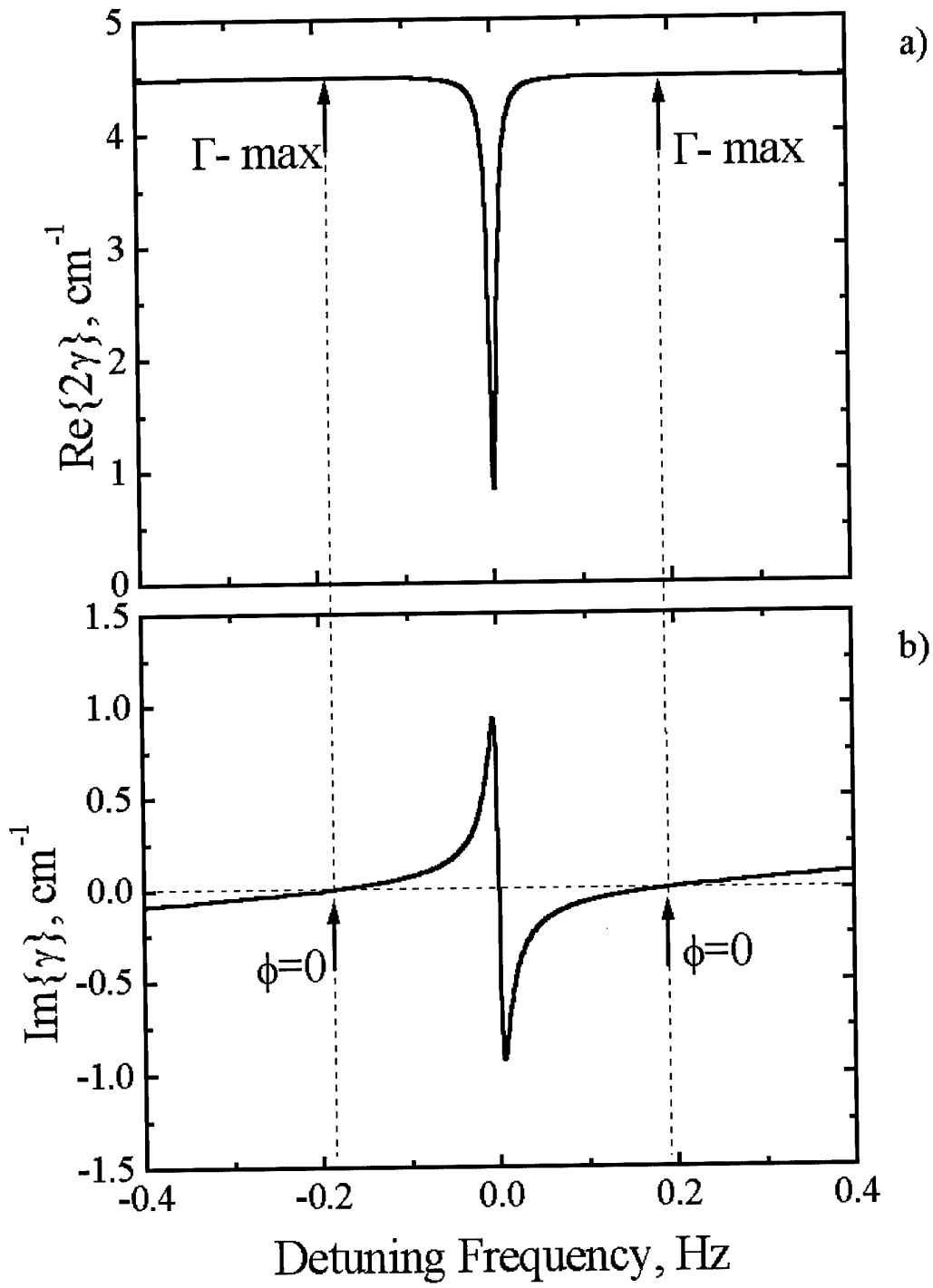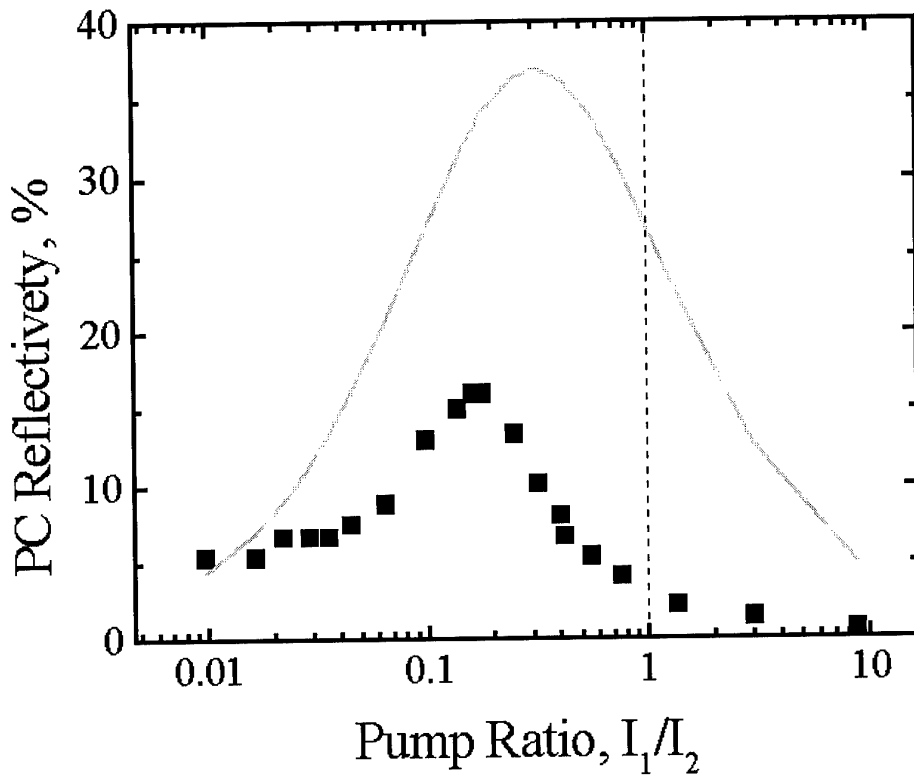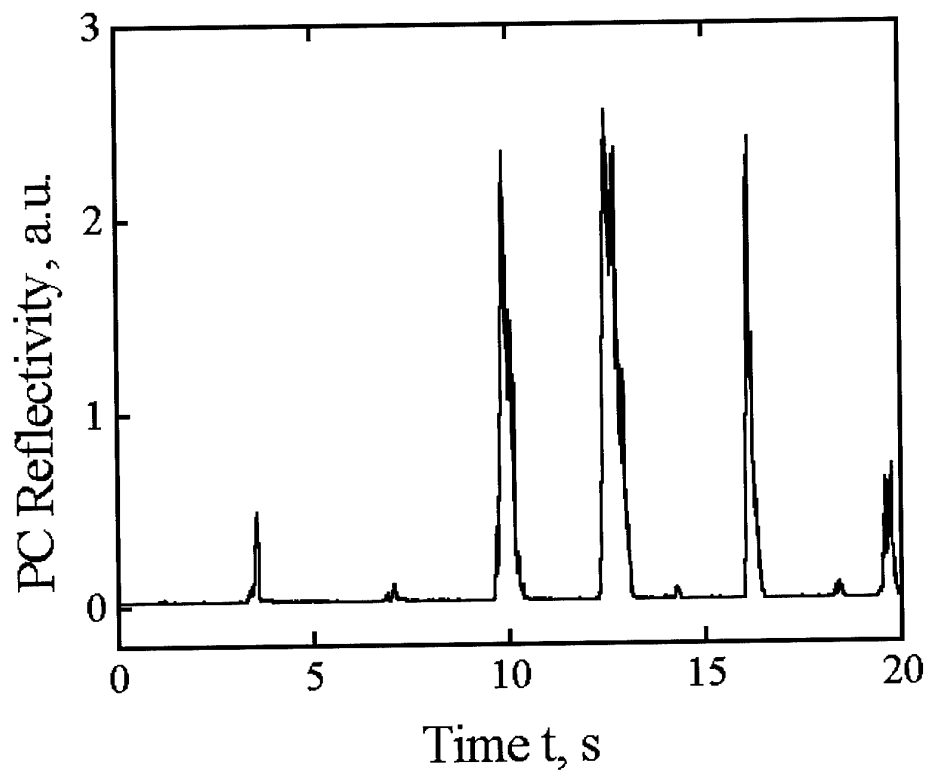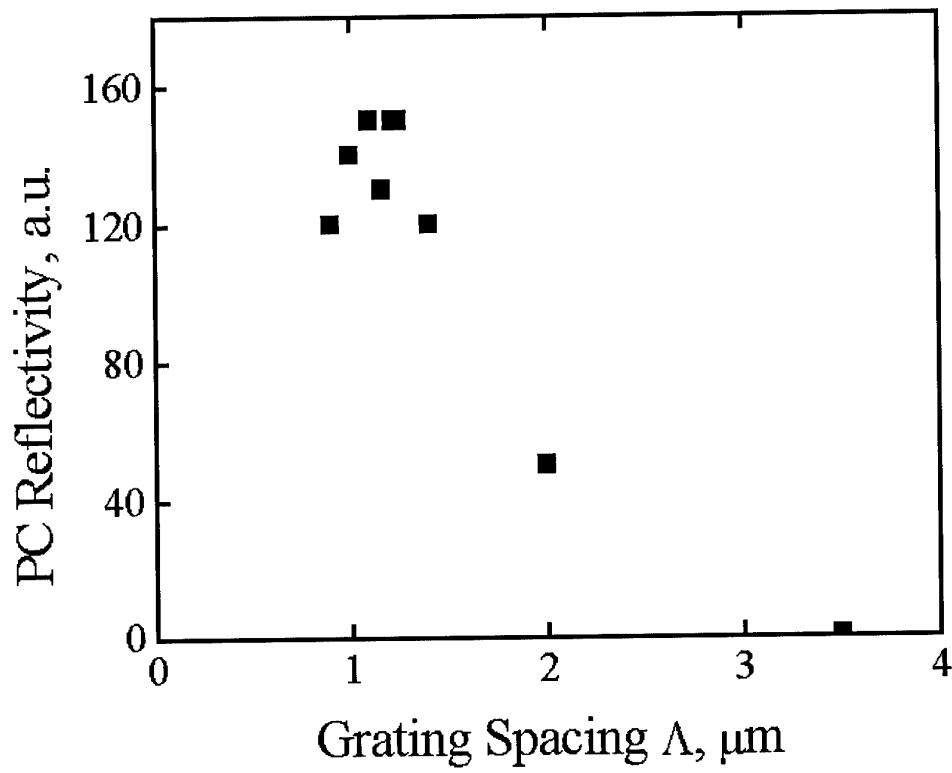3 D. D. Nolte, Photorefractive transport and multi-wave mixing, pp.1-67. Photorefractive effects and materials, Kluwer Academic Publishers. 1995.

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LIST OF PUBLICATIONS

During the period of the running Project two articles were published in Topical Issue of J. Opt. Soc. America on Photorefractive materials:

A. Shumelyuk, S. Odoulov, G. Brost "Nearly degenerate Two-Beam Coupling in photorefractive crystals with two types of movable species", J. Opt. Soc. Am. 37, N7, 2125-2131 (1998).

G. Brost, J. Norman, K. Shcherbin et al. "Study of photorefractive recording in semi-insulating semiconductors with nearly degenerate four wave mixing" J. Opt. Soc. Am. 37, N 7, 2083-2091 (1998).

two articles are prepared for publication:

A. Shumelyuk, S. Odoulov, G. Brost, and A.Grabar,
Spectral sensitivity of tin hypotiodiphosphate,

A. Shumelyuk, S. Odoulov, G. Brost, and A. Grabar,
Phase conjugation with photorefractive $Sn_2P_2S_6$.

and five talks were presented

A. Shumelyuk, S. Odoulov, G. Brost, Ute Hellwig, Yi Hu, and E. Kraetzig,
Photorefractive response of $Sn_2P_2S_6$ in wide spectral range from 0.63 to
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A. Shumelyuk, S. Odoulov et al, Anisotropy of photorefractive beam coupling in $Sn_2P_2S_6$, "Photorefractive Materials, Effects and Devices", Proceedings of 1977 Topical Meeting on "Photorefractive Materials, Effects and Devices", Chiba, June 1997, Published by Waseda University, Tokyo, pages 169-172.

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A. Shumelyuk, S. Odoulov, G. Brost, Ute Hellwig, Yi Hu, and E. Kraetzig, Ti-sapphire laser beamcoupling in $Sn_2P_2S_6$ CLEO'98, San-Francisco, May 1998, (CTuM58)

Attachment 1

PROPOSAL SUBMITTED TO EOARD

1. Title of Proposal: Optical Phase Conjugation with Photorefractive $Sn_2P_2S_6$

2. Date Submitted: May 5, 1997

3. Type of Proposal:

4. Requested Starting Date: July 1, 1997.

5. Proposed Duration: 12 months.

6. Total Amount Requested: 10,000 USD

7. Name of Organisation: Institute of Physics,   National Academy of Sciences of Ukraine, Kiev.

8. Type of Organisation: Institute.

9. Principal Investigator: Serguey Odoulov

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   fax: +380 (44) 256 1589
   e-mail: sodoulov@odoulov.kiev.ua
   odoulov@marion.iop.kiev.ua

10. Business Office: Serguey Odoulov
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   252 650 Kiev, Ukraine
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   fax: +380 (44) 256 1589

Introduction

Nonlinear Four-Wave Mixing in photorefractive materials allows for generation of phase-conjugate light waves at relatively low (down to microwatt) intensity levels. With the crystals known at present the best performance of Phase Conjugate Mirrors (PCM) was achieved in the visible region of spectrum. At the same time the phase conjugation in Near Infrared would be of great interest because of possible applications with well developed diode-pumped Nd^{3+}:YAG lasers. Laser-beam clean-up, image deblurring, optical correlation are only few examples of what can be done.

Several laboratories all over the world are working now in this field trying to solve the main problem which is the problem of suitable material. The known photorefractive semiconductors ensure quite fast response with poor gain factor while recently developed rhodium-doped barium titanate provides rather high gain with considerably longer response times (hundreds of seconds).

The purpose of this project is to use different wave-mixing processes in new photorefractive material, tin hypothiodiphosphate $(Sn_2P_2S_6)$ to generate the phase conjugate waves in the red and near infrared region of spectrum.

Our previous study of two-beam coupling in $Sn_2P_2S_6$, undertaken together with the group of Dr. George Brost from Rome Laboratory (see, e.g., Jap. J. Appl. Phys., v.35, part I, N 9B, pp.5154-5156, 1996, and Appl. Phys. Lett, v.69, N 24, pp.3665-3667, 1996) showed that this material combines relatively fast response (milliseconds range) with sufficiently large gain factor (7... 8 reciprocal centimeters). At the same time the presence of two out-of-phase photorefractive gratings in $Sn_2P_2S_6$ does not allow for trivial extension of the known techniques of phase conjugation to this material.

Description of Work

The main goal of the project is to get the largest possible phase conjugate reflectivity in $Sn_2P_2S_6$ by optimizing the usual parameters (as pump intensity ratio, signal to pump intensity ratio, angle of incidence for pump and probe waves, polarization of interacting waves, orientation of waves with respect to sample principal axes) and frequency shift, a new parameter of special importance for $Sn_2P_2S_6$.

Following work will be performed to achieve the abovementioned goal:

At first stage the backward wave four wave mixing will be used to generate the phase conjugate waves and optimum conditions will be found.

At the second stage we intend to study different self phase conjugate mirrors based on various coherent optical oscillators. As no seed radiation is required to produce the pump waves for these configurations, the frequency shift of the conjugate wave, we suppose, will be optimized automatically. One more time the optimum conditions for efficiency of phase conjugation will be found.

And to the end we hope to made operating the Double Phase Conjugating Mirror, to study it and to optimize its performance.

Equipment

Basic equipment (YAG:Nd^{3+} diode pumped laser, optical table, holders and optics, PC-controlled measuring devices) is available in our laboratory now. Therefore the main expenses will be related to Labor Costs and institutional Overheads. For successful work we will need also to replace our old Analog-Digital Converter Card and to install a new hard disk (or ZIP) with larger memory.

Qualification of Personnel

S.Odoulov:
Professor, Doctor of Science (University degree),
Head of Dynamic Holography Group.

K.Shcherbin:
Candidate of Science, (Technical University degree);
Senior Scientific Researcher.

A.Shumeljuk:
Candidate of Science (University degree),
Scientific Researcher.

Cost Estimate For Research Period
July 1, 1997 to June 30, 1998

1. Equipment purchased:
PC accessories
(AD Card and new hard disk) $ 600

2. Consumable,
Fax and e-mail expenses $ 200

3. Overhead charges: $ 2,000

4. Labor costs: $ 7,200

Total Estimated Costs $ 10,000

Schedule of Reports

The interim reports will be presented after two first stages of the Project (4 months and 8 months after beginning of the work); final report will be presented at the end of contract.

Principal Investigator

Serguey Odoulov

Interim Report
E O A R D Special Project SPC-97-4064
Contract F61708-97-W0206

During the first period of this work the calculations of the efficiency of nearly degenerate two-beam coupling were accomplished and presented in regular 1977 Topical Meeting on Photorefractive Materials, Effects and Devices, Chiba, June 1997 [1] and manuscript has been sent for publication [2].

According to the Workplan of present Project we were investigating the generation of the Phase Conjugate light beams in backward-wave four-wave mixing geometry. A He-Ne laser light (0.6328 μm, 40 mW output power) was used at this stage of the work.

At first, the characterization of a new SPS sample K3 (cut along the crystallographic directions and measuring 0.9 cm x 0.9 cm x 0.45 cm) has been performed. As this sample has all faces optically finished we were able to measure the two-beam-coupling gain factor for all possible orientations of grating vector with respect to crystallographic axes. It has been shown that the largest refractive index change is achieved for grating vector aligned along [100] direction when light waves are polarized nearly along the same direction [3]. The presence of two out-of-phase photorefractive grating was detected both with red and near-infrared light.

Next, a classical arrangement with two independent counterpropagating waves was used to generate the phase conjugate wave. A typical temporal dynamics of the phase conjugate wave is shown in Figure 1 [Fig.8a of Final Report]. A rather sharp increase of the phase conjugate reflectivity to approximately 10% is followed by slow decay to ten times smaller steady state value. This fall-off of efficiency is explained by the formation of supplementary space charge grating by moveable charge species of opposite sign and it is inevitable for this material with strictly degenerate four-wave mixing at ambient temperature.

To avoid the formation of the slow grating we check the possibility to build up a ring-loop coherent oscillator which is at the same time self-pumped phase conjugate mirror. In agreement with our expectations the "slow" grating do not appear in this case and the saturated value of phase conjugate reflectivity is nearly the same as its peak value (Fig.2) [Fig.12a of Final Report]. The pronounced beat frequency about 0.2 cycles/s is observed in output radiation, pointing to the inherent frequency shift of the generated phase conjugate wave (Fig.3) [Fig.13 of Final Report]. The phase conjugate reflectivity up to 16% is achieved for optimized fringe spacing about 3 μm. Taking into account that this value is measured directly and not corrected for rather high Fresnel reflection from the crystal input and output faces it looks quite acceptable.

The weak point of this work is poor quality of sample face polishing. First, it gives a lot of scattered light and limits the quality of phase conjugation. Second, the presence of scattered light reduces the ultimate achievable phase conjugate reflectivity. We are trying to solve this problem now using different polishing techniques.

Principal Investigator,

Serguey Odoulov

Manuscripts and Publications

1. S. Odoulov, A. Shumelyuk, and G. Brost. "Nearly Degenerate Two-Beam Coupling in $Sn_2P_2S_6$", Proceedings of 1977 Topical Meeting on Photorefractive Materials, Effects and Devices, Chiba, June 1997, Published by Waseda University, Tokyo, pages 173-176.

2. S. Odoulov, A. Shumelyuk, and G. Brost. "Nearly Degenerate Two-Beam Coupling in $Sn_2P_2S_6$", Submitted to JOSA B Topical Issue on Photorefractive crystals.

3. A. Shumelyuk, S. Odoulov et al. "Anisotropy of photorefractive beam coupling in $Sn_2P_2S_6$", Proceedings of 1977 Topical Meeting on Photorefractive Materials, Effects and Devices, Chiba, June 1997, Published by Waseda University, Tokyo, pages 169-172.

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