Double-phase conjugate mirror in nominally undoped $Sn_2P_2S_6$

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Coherent oscillation is achieved in the geometry of a double-phase conjugate mirror in nominally undoped $Sn_2P_2S_6$ with 647 nm radiation of a Kr^+ laser. The specific temporal dynamics of oscillation with modulated intensity and periodic $0-\pi-0-\pi$ variations of the oscillation wave phase is similar to that observed earlier using a semilinear oscillator geometry. The described oscillator ensures submillisecond delay time in the appearance of the phase conjugate wave. © 2009 Optical Society of America

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Tin hypothiodiphosphate ($Sn_2P_2S_6$, SPS) is a photorefractive material with fast response and relatively high gain [1]. Phase-conjugate waves have been generated with SPS in backward-wave four-wave mixing as well as with a ring-loop coherent oscillator [1]. During the past year two research teams reported on a double-phase conjugate mirror (DPCM) [2] in SPS that allows for simultaneous conjugation of two mutually incoherent light beams [3,4]. To get an efficient oscillation in the DPCM geometry with relatively high threshold (necessary coupling strength $\Gamma\ell \geqslant 4$), doped crystals, SPS:Sb [3] and SPS:Te [4], were used. A phase-conjugate reflectivity above 800% was achieved, e.g., with SPS:Te [4]. A more-modest efficiency was achieved with nominally undoped SPS, which was, however, sufficient to lock the emitters of a diode-laser bar to the master laser [4].

In this Letter we describe the use of nominally undoped SPS for mutual conjugation of 647 nm Kr^+ laser beams in a DPCM geometry. A particular feature of nominally pure SPS is the intensity dependence of the gain factor in a range of intensities where the photoconductivity is much stronger than the dark conductivity [1]. The use of Kr^+ laser light with an overall light intensity of $10\ \mathrm{W/cm^2}$ allowed us to reach a beam-coupling gain factor $\Gamma \geqslant 8\ \mathrm{cm^{-1}}$ and a phase-conjugate reflectivity $\geqslant 300\%$. Higher intensities also result in a remarkable decrease of the photorefractive grating buildup time, which goes down to the submillisecond range.

Apart from the practical aim of getting a fast-response phase-conjugate mirror, the study of an unusual temporal dynamics of coherent oscillation was the motivation of this work. The oscillation output shows periodic high-contrast variations of the intensity and a steplike variation of the phase by π between consecutive pulses. Following [6], we explain this behavior by a transient in-phase superposition of two space-charge gratings formed by moving charge carriers of opposite sign.

The experiments are performed with a nominally undoped $Sn_2P_2S_6$ sample (K3) that was grown in the Institute of Solid State Physics and Chemistry, Uzhgorod National University, Ukraine. The sample with dimensions $9 \times 4.5 \times 9\ \mathrm{mm^3}$ is cut along the crystallographic directions x, y, and z. The two-beam coupling gain factor in this sample is not the largest one for undoped crystals, which may indicate imperfect poling. The measured Γ was, however, independent of the particular position of the grating throughout the entire sample.

Light beams from an Ar^+-Kr^+ gas laser (800 mW in TEM_{00} mode at 647 nm, no etalon inside the cavity) enter the sample symmetrically through the z face so that the space-charge grating vector $\mathbf{K}$ is always aligned parallel to the x direction. With the polarization in the plane of incidence, [101], the coupling strength for recording of a transmission grating is optimized via the largest Pockel's tensor component r_{111} [1,5].

At first, two-beam coupling is studied in transmission grating geometry [Fig. 1(a)]. The dependence of the gain factor $\Gamma = (1/\ell)\ln(I_4/I_4^0)$ is measured versus the grating spacing $\Lambda = \lambda/(2\sin\theta)$ at different intensities of the pump wave $I_1 \gg I_4$. Here, I_4 and I_4^0 are the intensities of the transmitted signal wave with and without pump wave, respectively; ℓ is the sample thickness; and λ and θ are the wavelength and the beam crossing angle, both measured in air.

Figure 2 shows the increase of the peak value of the gain factor with the intensity up to $8\ \mathrm{cm^{-1}}$ at $10\ \mathrm{W/cm^2}$. The maximum coupling strength that the sample can ensure in our conditions $\Gamma\ell \approx 7$ is roughly two times larger than the smallest threshold value $\Gamma\ell_{th} = 4$ calculated for DPCM neglecting absorption [2]. The data of Fig. 2 also point to a quite obvious

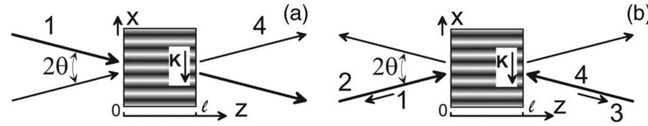

Fig. 1. Scheme of (a) beam coupling and (b) DPCM geometries. In (a) signal beam 4 is amplified at the expense of coherent pump beam 1. In (b) the path difference of beams 4 and 2, which impinge upon the sample, is larger than the laser coherence length, and new beams 3 and 1 are generated. The structure of the index grating is shown inside the samples.

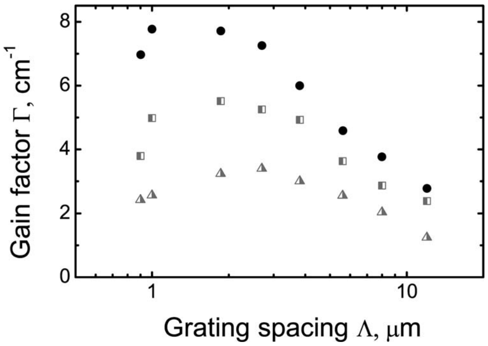

Fig. 2. Grating-spacing dependence of the gain factor Γ at 647 nm for intensities of the pump wave 0.1, 1.0, and 10 W/cm^2 (triangles, squares, and dots, respectively.)

decrease of the Debye screening length with increasing light intensity, thus supporting the hypothesis of a light-assisted modification of the effective trap density [1]. With a recording light intensity of 10 W/cm^2 the space-charge buildup time τ_f is below 200 μs at $\Lambda = 12$ μm.

Next, the two laser beams are loosely focused into the sample from the opposite z faces [Fig. 1(b)] with cylindric lenses of 115 mm focal length, forming a narrow luminous stripe in the plane of drawing, i.e., normal to the y direction. Special care is taken to ensure the best possible overlap of the two narrow beams in the bulk of the sample. With the intensity ratio $r = I_2/I_4 = 4$, oscillation is observed within the range of incidence angles θ that result in the formation of a shared transmission grating with spacings $4.5 \geqslant \Lambda \geqslant 0.9$ μm. The wavefronts of the generated beams are conjugate with respect to the wavefronts of the incident beams.

The initial stage of the oscillation buildup is similar to that of all other photorefractive oscillators; during a certain period of time the intensity of the oscillation wave is virtually zero, but then a nonlinear steplike increase of the intensity occurs [Fig. 3(a)]. This delay time, as one can see from Fig. 3(a), can be smaller than 1 ms at a total light intensity of 10 W/cm^2.

An unusual feature of the oscillator under study consists of periodic nonharmonic variations of the oscillation intensity for exposure times exceeding the dielectric relaxation time by many orders of magnitude [Fig. 3(b)]. It was established, with the interferometric technique [6], that for each new oscillation pulse from the sequence shown in Fig. 3(b) the phase of the oscillation wave differs from the phase of the previous pulse by π.

The dependence of the phase-conjugate reflectivity $R_{pc} = I_3(\ell)/I_4(\ell)$ on the intensity ratio of the incident beams $r = I_2(0)/I_4(\ell)$ [Fig. 4(a)] is typical for photorefractive DPCMs [2]. The threshold values of r, beyond which the oscillation vanishes, allow for evaluation of the ultimate coupling strength of our sample via the relationship [2]

$$\Gamma \ell_{th} = 2\left(\frac{r+1}{r-1}\right)\ln r. \quad (1)$$

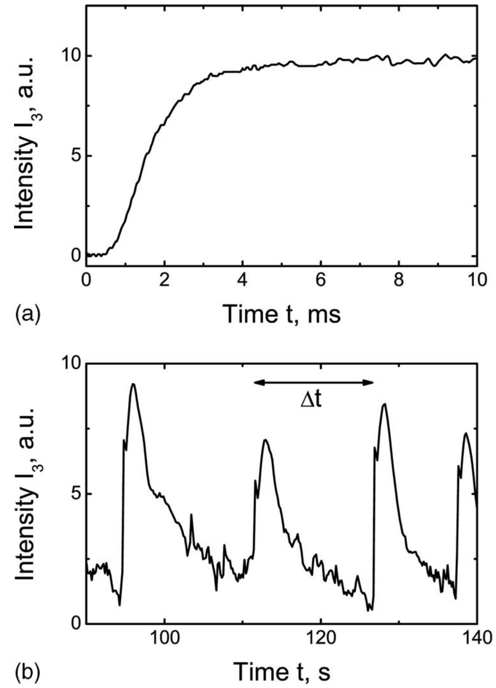

Fig. 3. Temporal dynamics of the oscillation intensity (a) just after exposure of a virgin sample and (b) for well-established oscillation ($\Lambda = 1.4$ μm, $r = 4$). Note the difference in the time scale for the two frames.

The oscillation disappers at $r_1 \approx 0.022$ and $r_2 \approx 45$ [Fig. 4(a)]. For these pump ratios the value $\Gamma \ell \simeq 8$ is determined via Eq. (1), which is close (within 20% error bars) to that directly measured from the beam-coupling experiment, $\Gamma \ell \simeq 7$.

The data on the coupling strength can also be extracted from the range of grating spacing within which the oscillation exists. The values of Λ at which the oscillation disappears allow us to estimate the

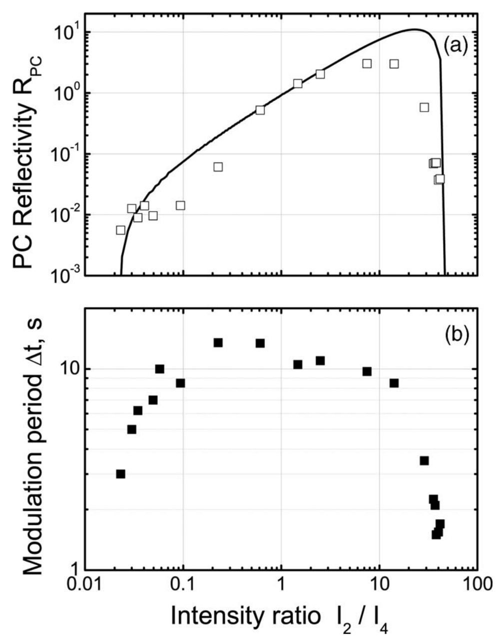

Fig. 4. (a) Phase-conjugate reflectivity and (b) period of intensity variation versus beam ratio for $\Lambda = 1.4$ μm. The solid line is the dependence calculated in plane-wave approximation [2] for lossless medium and $\Gamma \ell = 8$.

threshold coupling strength from the dependence of Fig. 2 for 10 W/cm^2. With the ultimate coupling strength of about $\Gamma\ell \approx 7$, the threshold coupling strength evaluated from Fig. 2 for $\Lambda = 4.5\ \mu$m is $\Gamma\ell_{th} \approx 4.9$. This value is in reasonable agreement with $\Gamma\ell_{th} = 4.6$, which follows from Eq. (1) for $r = 4$.

Figure 4(b) shows the dependence of a temporal modulation period Δt in the oscillation intensity [see Fig. 3(b)] on intensity ratio of the incident beams. This period Δt decreases by 1 order of magnitude at pump ratios r_1 and r_2, i.e., in the vicinity of the oscillation threshold.

The dependences shown in Figs. 3(b) and 4(b) are similar to that reported for SPS in a semilinear oscillator in [6]. Thus, DPCM with nominally undoped $Sn_2P_2S_6$ is a second type of coherent oscillator that features this quite-unusual temporal dynamics. Its origin is most probably related to the presence of two types of movable charge carriers in this material, presumably photoexcited holes [7] and thermally excited electrons. The hole conductivity in $Sn_2P_2S_6$ is much stronger than the electron conductivity, so that the two dielectric relaxation times differ by several orders of magnitude. As a consequence, the system shows a fast transient gain that is followed by a slower compensation and a much more modest coupling in the steady state [1].

Qualitatively, the oscillation dynamics can be explained as follows. At first, after the exposure of a virgin crystal to the light intensity, the fast grating starts to develop and the two-beam coupling gain increases until the threshold condition of oscillation [Eq. (1)] is fulfilled. The amplitude of the slow grating at this moment is negligibly small because of the big difference in characteristic times. This is why all threshold characteristics depend solely on parameters of the fast grating. This means that all estimates for $\Gamma\ell$ given above are related to the fast grating. With exposure times that are still much shorter than the slow time, the dynamics of oscillation does not differ from that for DPCM with photorefractive crystals that feature monopolar conductivity [see Fig. 3(a)].

For longer exposure times, the amplitude of the slow grating, which is π shifted in phase with respect to the initial fast grating, increases and the overall gain becomes smaller until the threshold of oscillation is reached. Consequently, the oscillation intensity disappears at the threshold. With zero oscillation intensity the fast grating vanishes within a few milliseconds, but the slow grating remains virtually the same. The wave diffracted from this grating serves as a seed for the recording of a new fast grating that now is in phase with the slow one. When the oscillation intensity reaches its maximum value, the electrons start to redistribute and rebuild the slow grating to compensate the existing space-charge grating; the entire cycle is then repeated [6]. Thus the duration of a pulse in the oscillation intensity depends on the slow decay time of the system and on the difference of the sample coupling strength and the threshold coupling strength. The closer the coupling strength is to its threshold value, the shorter time is necessary to destroy the oscillation and the smaller Δt becomes [Fig. 4(b)].

To summarize, we report on a DPCM with submillisecond switch-on time and a reflectivity exceeding 300% at 647 nm. In the saturated regime, the reflected beam features a high-contrast intensity and phase modulation that is the consequence of competition of the space-charge gratings formed by two movable charge species of different signs.

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