Transient gain enhancement in photorefractive crystals with two types of movable charge carrier

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Considerable improvement of a transient two-beam coupling gain is reported for $Sn_2P_2S_6$, a photorefractive crystal that possesses two types of movable charge carrier. A gain enhancement occurs if the phase difference of the interacting beams is abruptly changed to π. Enhancement is also achieved with periodic phase variations of zero and π between two discrete states at modulation frequencies lower than the smallest of two reciprocal characteristic times of the space-charge formation. © 2007 Optical Society of America

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The presence of secondary charge carriers in crystals usually inhibits the steady-state photorefractive response. Whatever the origin (thermally excited or photoexcited, electrons–holes, or moving ions) the secondary charges tend to compensate for the space-charge grating created by the principal photoexcited carriers. This results in a reduction of the overall space-charge field and therefore in a smaller index modulation via the Pockels effect [1,2].

The undesirable effect of grating compensation can be overcome if frequency detuning is introduced in one of two interacting waves. The slow-response grating is more strongly affected by the fringe motion than is the fast-response grating; therefore it is possible to find an appropriate detuning where the slow grating is practically suppressed while the fast grating still has nearly the same amplitude as in the degenerate case. This technique of gain enhancement was successfully used in Ref. [3] with $Sn_2P_2S_6$, the photorefractive crystal that ensures large gain ($10\ cm^{-1}$) and a millisecond response time [2].

The space-charge gratings formed by the carriers of different signs are most often π shifted with respect to each other, which explains the inhibition of the photorefractive response. The exception to this rule is so-called temperature-intensity resonance in semiconductors [4], where quite large electric fields partially destroy the unfavorable, out-of-phase superposition of the space-charge gratings. A steady-state gain factor of up to $11\ cm^{-1}$ has been achieved with this technique in InP:Fe.

We present in this Letter experimental evidence of a transient in-phase superposition of the space-charge gratings that becomes possible after an abrupt change of the phase difference between the interacting waves to π. The transient gain that occurs after the steplike phase modulation appears to be higher as compared with not only the steady-state gain in the degenerate case, but also the steady-state gain at optimized frequency detuning.

To measure the two-beam coupling gain the standard transmission grating geometry is used. Two beams from a He–Ne laser (633 nm), polarized in the plane of incidence, impinge upon the Z-cut $Sn_2P_2S_6$ crystal so that the fringe wave vector is aligned along the OX axis. The signal-to-pump intensity ratio is 1:20,000. The nominally undoped sample K3 (with thickness $l=9$ mm) reveals the pronounced competition of two out-of-phase gratings in beam coupling dynamics: with the pump wave switched on at $t=0$, the output intensity of the signal wave increases more than 250 times within several milliseconds and then decreases with a much slower rate. This behavior is shown as a first peak (time t up to 75 s) in Fig. 1.

At $t=75$ s, when the steady-state is practically established, the phase shift π is introduced in the input signal wave via fast displacement of a piezo-mounted mirror. A new pulse develops with the same buildup and decay rates, but with an obviously higher peak intensity as compared with the first peak. If the phase shift π is introduced after the steady-state is reached again, the next pulse is identical to the previous. The pulse sequence shown in Fig. 1 is registered for a grating spacing $\Lambda=0.9\ \mu$m. The initial peak amplitude shown in Fig. 1 arises from the optically excited diffusion grating alone. The subsequent peaks are greater in amplitude because they include additional coupling from the second grating, which

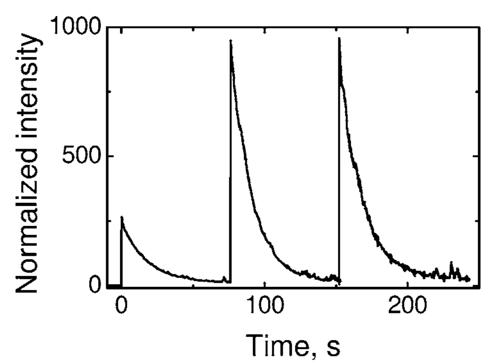

Fig. 1. Temporal dependence of the output signal intensity normalized to the signal intensity with no pump wave. At $t=75$ s and $t=150$ s the phase of the input signal wave is changed to π.

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arises from space-charge-field-induced drift and segregation of optically inactive moveable carriers.

In the next experiment a periodic phase alternation 0-π-0-π-0- is introduced into the input signal wave, and the transient gain factor $\Gamma^{tr} = (1/\ell)\ln(I^{max}/I^{in})$ is measured as a function of the phase modulation frequency Ω. Here I^{max} and I^{in} are the peak pulse intensity and the cw output signal wave intensity with the pump wave switched off, respectively. The measured dependence for $\Gamma^{tr}(\Omega)$ for $\Lambda = 0.9\ \mu$m is shown in Fig. 2. For comparison, we show the frequency dependence of the steady-state gain factor $\Gamma^{ss} = (1/\ell)\ln(I^{out}/I^{in})$ for nearly degenerate two-beam coupling measured for our sample as described in Ref. [3]. Here I^{out} is the output intensity of the amplified signal wave, and I^{in} is still the transmitted signal wave intensity with the pump wave switched off. The frequency detuning is introduced with the help of the same piezo-mounted mirror, via sawtooth modulation of the signal wave phase. The amplitude of this phase modulation is set to be 2π; the frequency detuning in this case is equal to the sawtooth modulation frequency.

It is obvious that at large modulation frequencies ($\Omega \geq 1$ Hz) the two dependences practically coincide. At small modulation frequencies they differ both quantitatively and qualitatively: while the steady-state gain factor Γ^{ss} drops to quite a low level with decreasing Ω, the transient gain factor Γ^{tr}, in contrast, increases and saturates at a level that can never be reached by the steady-state gain. We attribute this enhancement to the transient in-phase superposition of two space-charge gratings formed by movable charge carriers of different signs.

Qualitatively the dynamics of beam coupling with deep phase modulation can be described as follows. When a virgin sample is illuminated by two recording waves, the fast grating develops within several milliseconds. Being $\pi/2$ shifted in phase with respect to the fringes, this grating ensures unidirectional beam coupling, and the weak signal beam gains intensity at the expense of the strong pump wave [2].

The carriers responsible for the fast grating formation are photoexcited holes [5]. In addition, a certain number of optically inactive movable charge carriers of another type exist in nominally pure $Sn_2P_2S_6$ crystals. These carriers move in a space-charge field of the fast grating to form the compensating out-of-phase grating. The buildup time of this grating is much longer than the buildup of the fast grating; this explains the slow decrease of the signal wave intensity after the transient peak.

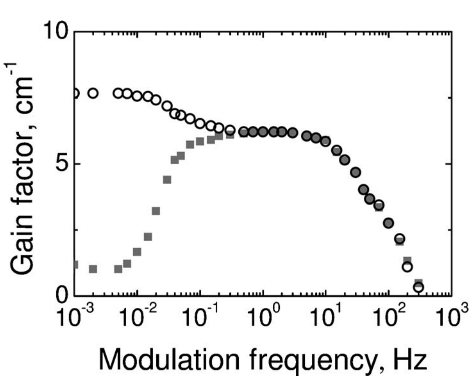

Fig. 2. Modulation frequency dependence of the transient gain factor (open circles) for periodic 0-π-0-π-0-π- phase modulation and the steady-state gain factor (gray squares) for sawtooth phase modulation.

Let us now consider the effect of phase modulation. If after steady-state is reached the phase difference of the interacting waves is changed to π, the contrast of the light fringes in the crystal is reversed (fringes are moved laterally to half of the fringe spacing). The fast component of the grating is no longer matched to the new position of the fringes; therefore it decays to zero and reappears shifted by π with respect to its initial position.

All these changes take place within several tens of milliseconds; during this time the slow grating does not change in amplitude or position. In such a way the new recorded fast grating appears to be in phase with the slow grating that remains from the previous pulse. This situation is, however, not equilibrium: the thermally generated carriers start to move to compensate for the newly developed space-charge field. At first they destroy the in-phase slow grating and then build a new one that is out of phase with the fast grating. Thus the conventional steady-state with small differential gain is reestablished. The next abrupt π change of the phase difference triggers exactly the same sequence of events: quick erasure of the out-of-phase fast grating, quick recording of the new in-phase fast grating, slow erasure of the in-phase slow grating, and slow buildup of the out-of-phase slow grating until saturation is reached or until the next abrupt π change of the phase difference. For periodic π–zero phase modulation this process occurs repeatedly.

The largest transient gain is achieved if the interval between two consecutive variations of phase T is longer than the buildup time of the slow grating τ_s (or the modulation frequency is smaller than the slow grating decay rate, $\Omega \leq 1/\tau_s$). If the period T becomes much smaller than τ_s, the slow grating does not develop, and no gain enhancement is observed (Fig. 2). The measured transient gain is ensured by the fast grating only. For T comparable with the buildup time of the fast grating, even the fast grating does not develop in full; therefore the transient gain decreases with increasing modulation frequency.

We experimentally show and qualitatively explain the enhancement of the transient gain factor at low modulation frequencies. A question may arise about the origin of such an enhancement. In photorefractive crystals the ultimate gain factor is limited by the electro-optic properties of the crystal and the built-in space-charge field (see, e.g., Ref. [1]). In turn, for diffusion-driven recording, the space-charge field E_{sc} cannot be larger than the diffusion field, $E_D = k_b TK/e$. If with only one fast grating E_{sc} reaches its upper limit imposed by the diffusion field, by no means is it possible to increase it further, regardless of the number of different types of carrier involved in the grating formation. This restriction does not apply, however, in the case of a trap-density-limited space-

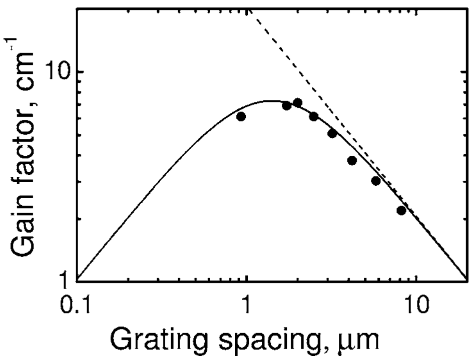

Fig. 3. Grating spacing dependence of the fast grating gain factor (circles). Best fit of calculated dependence with the space-charge limitation taken into account (solid curve). Expected dependence for infinite effective trap density (dashed line).

charge field, which is typical for $Sn_2P_2S_6$. In the case of trap density limitation for the fast grating, the in-phase addition of the slow grating may and should enhance the overall space-charge field and increase the gain factor.

In Fig. 3 we plot the grating spacing dependence of the gain factor for the fast grating in our $Sn_2P_2S_6$ sample. As one can see, the gain factor has a well-defined maximum near $\Lambda = 1.5\ \mu$m, thus proving serious trap density limitations for gratings with Λ smaller than this value. It should be noted that all measurements described above are done at $\Lambda = 0.9\ \mu$m, i.e., in the domain with pronounced space-charge limitation.

To prove that the space-charge limitation is a necessary condition to achieve the enhancement of gain, we perform the measurements of beam coupling dynamics similar to that shown in Fig. 1 at $\Lambda = 8\ \mu$m. No gain enhancement is observed within the experimental accuracy, in accordance with our expectations. Thus we conclude that the proposed technique is efficient for the interaction of two beams that produce photorefractive gratings with high spatial frequencies, including the interaction of counterpropagating waves.

The intuitive explanation presented above might suggest roughly an enhancement of the gain factor by two times when the initially out-of-phase gratings overlap in phase. In the experiment we observe, however, only 33% enhancement. The reason might be in the difference of the effective trap densities for different charge species. The quantitative description of this effect will be done within the standard models for crystals with two types of charge species (as in Ref. [6]) appropriately adapted for $Sn_2P_2S_6$ [7].

The results obtained are also direct proof of the model that explains the nontrivial temporal dynamics of a $Sn_2P_2S_6$-based coherent oscillator in a semilinear cavity [8]. This oscillator generates periodic sequences of nearly triangular pulses with a spontaneous (not imposed from outside) phase change to π for every newly generated pulse with respect to the previous one. The advantage of such an operation mode, as compared with cw oscillation at a shifted frequency, is simply related to the higher reported transient gain compared with the maximum achievable steady-state gain.

One possible application of the considered technique consists of the transformation of a cw input wave into a sequence of pulses with a peak intensity much higher than the intensity of the input wave. Depending on how the phase difference is introduced (the phase of either wave can be modulated), the phase of all pulses in sequence is the same or may change alternatively between two discrete values that differ by π.

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