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**TRANSIENT GAIN ENHANCEMENT IN
PHOTOREFRACTIVE CRYSTALS WITH TWO TYPES OF
MOVABLE CHARGE CARRIERS (PREPRINT)**

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**Hardened Materials Branch
Survivability and Sensor Materials Division**

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14. ABSTRACT Considerable improvement of a transient two-beam coupling gain is reported for Sn ₂ P ₂ S ₆ , a photorefractive crystal that possesses two types of movable charge carriers. A gain enhancement occurs if the phase difference of the interacting beams is abruptly changed to π . It is also achieved with periodic phase variations between two discrete states, zero and π , at modulation frequencies less than the smallest of two reciprocal characteristic times of space charge formation. The direct observation of gain enhancement, which is due to forced deep phase modulation, clarifies the origin of nontrivial dynamics of the coherent oscillation in Sn ₂ P ₂ S ₆ called "optical multivibrator."					
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Transient gain enhancement in photorefractive crystals with two types of movable charge carriers

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Considerable improvement of a transient two beam coupling gain is reported for $\text{Sn}_2\text{P}_2\text{S}_6$, a photorefractive crystal that possesses two types of movable charge carriers. A gain enhancement occurs if the phase difference of the interacting beams is abruptly changed to π . It is also achieved with the periodic phase variation between two discrete states zero and π at modulation frequencies less than smallest of two reciprocal characteristic times of the space charge formation. The direct observation of gain enhancement, which is due to forced deep phase modulation, clarifies the origin of nontrivial dynamics of the coherent oscillation in $\text{Sn}_2\text{P}_2\text{S}_6$ called "optical multivibrator".

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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 photo excited, electrons/holes or moving ions) the secondary charges tend to compensate the space charge grating created by the principal (main) photoexcited carriers. This result in reduction of overall space charge field and therefore in smaller index modulation via Pockels effect.^{1,2} Reducing the space charge we reduce in chain the index modulation, grating efficiency, beam coupling.

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 much stronger affected by the fringe motion than 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 for degenerate case. This technique of gain enhancement was successfully used with $\text{Sn}_2\text{P}_2\text{S}_6$ in Ref. 3.

The space charge gratings formed by the carriers of different sign are most often π shifted with respect to each other, which explains the inhibition of photorefractive response. 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 step-like phase modulation appears to be higher not only as compared to the steady-state gain in the degenerate case, but also the steady-state gain at optimized frequency detuning. The periodic deep phase modulation $0, \pi$ allows for pulse trains with much higher peak intensity (nearly 1000 times) than the intensity of the cw input signal wave. Depending on how the phase difference is introduced (whether the phase of the pump or signal wave is modulated) the consecutive pulses in train will have either the same phase, or will differ in phase by π . The peak intensity of these pulses is much higher (nearly 1000 times) than the intensity of the cw input signal wave. To measure the two-beam-coupling gain the standard transmission grating geometry is used. Two beams from a He-Ne laser polarized in the plane of incidence, impinge symmetrically upon the Z-cut $\text{Sn}_2\text{P}_2\text{S}_6$ crystal so that the fringe wavevector is aligned along OX axis. The signal-to-pump intensity ratio is 1:20,000. The nominally undoped sample K3 (with thickness $\ell = 9$ mm) reveals 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 rapidly increases more than 250 times within several milliseconds and then gradually decreases with a much slower rate. This behavior is shown as a first peak (time t from 0 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 mirror mounted on piezoceramic. A new pulse develops with the same build-up and decay rates, but with an obviously higher peak intensity as compared to the first peak. If the phase shift π is introduced after the steady-state is reached again, the next pulse is identical to previous, i.e., it has a much higher intensity than the very first pulse at $t = 0$. The pulse sequence shown in Fig.1 is registered for grating spacing $\Lambda = 0.9 \mu\text{m}$. The initial peak amplitude shown in figure 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 arises from space-charge field induced drift and segregation of optically inactive moveable carriers.

In the next experiment a periodic phase alternation $0-\pi-0-\pi-0-$ is introduced into the input signal wave and the transient gain factor

$$\Gamma^{tr} = (1/\ell)\ln(I^{max}/I^{in}) \quad (1)$$

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. Figure 2 represents the measured dependence $\Gamma^{tr}(\Omega)$ for grating spacing $\Lambda = 0.9 \mu\text{m}$, the same as in previous experiment. For comparison, we show in the same figure the frequency dependence of the steady-state gain factor

$$\Gamma^{ss} = (1/\ell)\ln(I^{out}/I^{in}) \quad (2)$$

for nearly degenerate two-beam coupling measured for our sample as it is described in Ref. 3. Here I^{out} is the output intensity of the amplified signal wave with 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 saw-tooth 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 saw tooth modulation frequency.

It is obvious that at large modulation frequencies ($\Omega \geq 1 \text{ Hz}$) 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} , on the contrary, increases and saturates at the level that can never be reached by the steady-state gain. We interpret this gain enhancement as a consequence of transient in-phase superposition of two space charge gratings formed by movable charge carriers of different sign.

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 formation of the fast grating are photoexcited holes.⁴ In addition, a certain amount of optically inactive movable charge carriers of another type exists in nominally pure $\text{Sn}_2\text{P}_2\text{S}_6$ crystals. These carriers move in a space charge field of the fast grating to form the compensating out-of-phase grating. The build-up time of the compensating grating is much longer than the build-up of the fast grating; this explains the slow decrease of the signal wave intensity after the transient peak.

Let us now consider the effect of deep phase modulation. If after steady-state is reached the phase difference of the interacting waves is suddenly changed to π , the contrast of the interference fringes in the crystal is reversed (fringes are moved laterally to a half of the fringe spacing). The fast component of grating is no longer matched to the new position of 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 millisecond; during this time the slow grating does not change in amplitude or position. In such a way the new recorded fast grat-

ing 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 new developed space charge field. At first they destroy the in-phase slow grating and then build a new one which is out-of-phase to 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 new in-phase fast grating, slow erasure of in-phase slow grating, slow build-up of out-of-phase slow grating until the 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 characteristic build-up time of the slow grating τ_s (or 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 that ensured by the fast grating only. For T comparable to the build-up 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. The 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.¹). In turn, for the diffusion-driven recording the space charge field E_{sc} cannot be larger than the diffusion field, $E_D = k_bTK/e$. 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 the number of different types of carriers involved in the grating formation. This restriction does not apply however in the case of a trap-density-limited space charge field, which is typical for $\text{Sn}_2\text{P}_2\text{S}_6$. In the case of trap density limitation for the fast grating, the in-phase addition of 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 two-beam coupling gain factor for the fast grating in our $\text{Sn}_2\text{P}_2\text{S}_6$ sample. As one can see, the gain factor has a well defined maximum near $\Lambda = 1.5 \mu\text{m}$ thus proving serious trap density limitations for gratings with smaller than this value. It should be noted that all measurements described above are done at $\Lambda = 0.9 \mu\text{m}$, i.e., in the domain with pronounced space charge limitation.

To prove that the space charge limitation is a necessary condition for implementation of the proposed technique of gain enhancement, we perform the measurements of beam coupling dynamics similar to that shown in Fig.1 at $\Lambda = 8 \mu\text{m}$. No gain enhancement is observed

within the experimental accuracy, in accordance to our expectations. In a graph similar to that of Fig.1 the amplitude of the second pulse (after switching the phase difference to π) appears to be the same as the amplitude of the initial pulse.

Thus we conclude that the proposed technique of gain enhancement is efficient for the interaction of two beams that produce photorefractive gratings with rather high spatial frequencies, including the interaction of counter-propagating waves. The results obtained are also direct proof of the model proposed for the explanation of nontrivial temporal dynamics of $\text{Sn}_2\text{P}_2\text{S}_6$ coherent oscillator in a semilinear cavity.⁵ 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 to cw oscillation at a shifted frequency, is simply related to the higher reported transient gain compared to maximum achievable steady-state gain.

One possible application of the considered technique consists in the transformation of a cw input wave into a sequence of pulses with the 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 can be one and the same or may change alternatively between two discrete values which differ to π .

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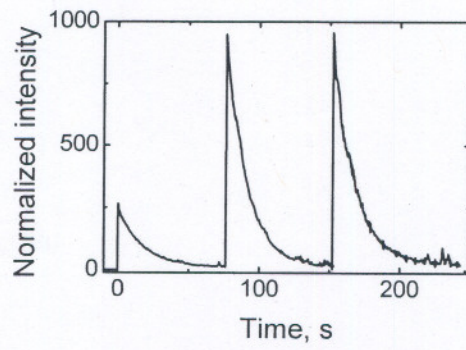


Fig. 1. Temporal dependence of the output signal intensity normalized to the signal intensity with no pump wave. At $t = 75$ s and 150 s the phase of the input signal wave is changed to π .

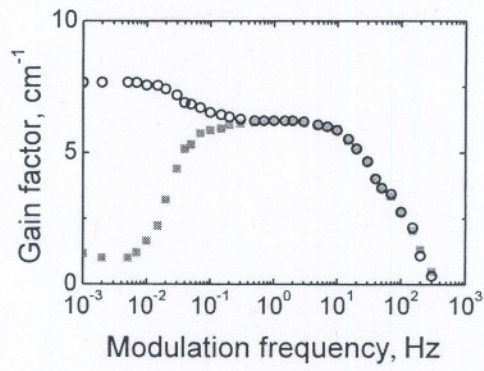


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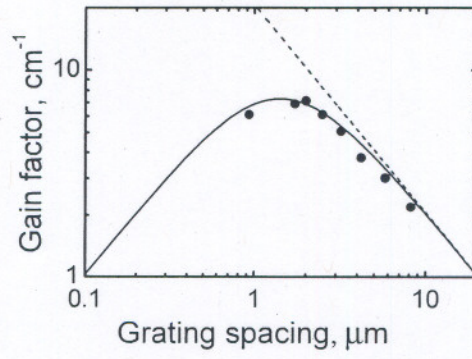


Fig. 3. Grating spacing dependence of the fast grating gain factor (circles). The best fit of calculated dependence with the space charge limitation taken into account (solid line). Expected dependence for infinite effective trap density (dashed line).