Slowing Down of Light in Photorefractive Crystals with Beam Intensity Coupling Reduced to Zero

A. Shumelyuk,[1] K. Shcherbin,[1] S. Odoulov,[1] B. Sturman,[2] E. Podivilov,[2] and K. Buse[3]
[1]*Institute of Physics, National Academy of Sciences, 252650 Kiev, Ukraine*
[2]*Institute of Automation and Electrometry, 630090 Novosibirsk, Russia*
[3]*Physical Institute, University Bonn, D-53115 Bonn, Germany*
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A considerable deceleration of light pulses in crystals with nearly compensated space-charge field proves that the strong dispersion of dynamic photorefractive gratings in the vicinity of Bragg resonance is of primary importance for light slowing down.

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Propagation of light pulses through a resonant system was already a topic over 100 years ago [1], but the last decade has brought a renaissance of interest to this field, especially after revealing the possibility of enormous deceleration, trapping, and coherent storage of light with electromagnetically induced transparency (EIT) [2]. This interest is related both to fundamental aspects of nontrivial light pulse manipulation and to possible applications for optical delay lines, for quantum computing, and for developing sensitive measurement techniques.

Recently it has been shown that another nonlinear effect, recording of dynamic refractive index gratings in photorefractive crystals, can be also used for the slowing down of pulse propagation [3,4]: a pulse velocity as low as 0.02 cm/s has been reported [3]. To get the slowing down of light pulses, crystals with a so-called nonlocal nonlinearity were used ($BaTiO_3$, $Sn_2P_2S_6$) that ensure a strong intensity coupling of two waves during the recording of a dynamic grating (see, e.g., [5]). Two waves were directed to the photorefractive crystal, the intense cw pump wave and the much weaker signal wave with Gaussian temporal envelope (Fig. 1). These waves recorded a dynamic refractive index grating in the sample, and, because of self-diffraction from this grating, the signal pulse was delayed, usually to a time comparable to the grating decay time in the crystal [3]. Simultaneously, the delayed pulse was strongly amplified ($\gg 10^3$ times) at the expense of the intensity of a strong pump wave.

The huge amplification of the signal pulse yields a simple qualitative explanation of the time delay of the output pulse: The initial part of the pulse is propagating in a medium with relatively weak refractive index grating; it is not strongly affected at the output. With progressing time, the amplitude of the index grating is increasing, and the number of photons diffracted from the pump wave into the signal is growing. This results in much stronger amplification of the falling-down pulse front as compared to the uprising front of the pulse. As a consequence, the maximum of the transmitted amplified pulse is delayed with respect to the maximum of the pulse that propagates through the sample with no pump wave. Not calling into question this intuitive explanation, we intend to emphasize that it is not the amplification itself which is crucially important for light slowing down with photorefractive gratings, but a large frequency dispersion of a propagation constant $dk/d\Omega$ that is typical for 3D dynamic gratings with long decay times (here k and Ω are the signal wave number and signal wave frequency detuning to the pump wave, respectively). Both effects, the gain and the nonlinear change of the phase, can be understood from the expression for steady-state output signal field, $A_s(\ell) = A_s(0)\exp(\gamma\ell)$, calculated in undepleted pump approximation [5]. Here γ is the coupling constant that depends on frequency detuning Ω of the signal wave to pump wave; it possesses both a real part responsible for amplification and an imaginary part that modifies a propagation constant k [5].

Just the strong dispersion mentioned above ensures a dramatic decrease of the light group velocity [1] in a similar way as for other narrow resonances like those related, e.g., to excitons in semiconductors [6], to EIT in gases [2], to a narrow quantum coherence "hole" in the homogeneously broadened absorption line of ruby [7,8].

For a majority of photorefractive crystals a large dispersion is inseparable from a large gain factor and the formulated statement about priority of dispersion might look senseless. It is possible to find, however, special conditions when large dispersion exists with virtually zero gain. To prove the primary importance of the dis-

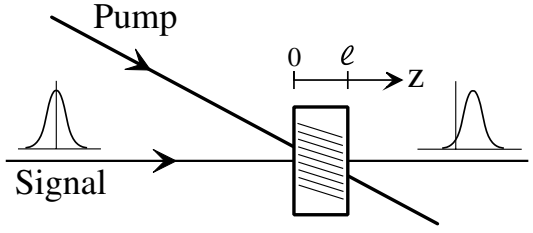

FIG. 1. Schematic of pulse propagation through a photorefractive crystal in the presence of a coherent pump wave.

persion we investigate the pulse propagation in photorefractive crystals with two types of movable species, one positively and the other negatively charged. With no external field applied to the sample and with identical temporal frequencies of the recording waves these charges form two out-of-phase gratings that can totally compensate each other and reduce to zero the intensity coupling of the signal wave to the pump wave. This complete compensation occurs, however, only in the frequency degenerate case. In crystals with strongly different decay times of two gratings even a slight deviation from the resonance (for 10^{-3} Hz in $Sn_2P_2S_6$ crystals [9]) affects one of the two gratings much stronger than the other, and the differential gain appears. This narrow resonance provides also a strong frequency-dependent change of the signal wave phase [9], i.e., leads to $k(\Omega)$.

It is shown that in a 9-mm-thick crystal that ensures very modest intensity coupling in the steady state (signal beam is depleted 1.5 times instead of being amplified $\simeq$ 10 000 times as in [3]) the signal pulse still can be delayed for several seconds. The analogy of grating-assisted light slowing down to that which is due to electromagnetically induced transparency, as is suggested in our previous Letter [3], becomes even more convincing with this result. In this Letter we are not looking for further improvement of already reported data on pulse velocity in photorefractive crystals (what can be done for sure with long-response-time materials); the aim is to prove a fundamental thesis that a considerable slowing down can be achieved in a medium with negligibly small beam coupling.

In what follows it is shown experimentally that a large dispersion can be achieved with virtually zero gain when taking as an example nominally undoped photorefractive tin hypothiodiphosphate ($Sn_2P_2S_6$, SPS) [10] and studying nearly degenerate coupling of two cw plane waves. While a gain spectrum with a hole has already been reported for SPS [9], the phase delay spectrum is measured in such a system for the first time. Then the data on light slowing down are presented, with gratings that are recorded in the same sample by two matched pulses, i.e., pulses possessing identical Gaussian temporal profiles. Finally we show how this technique of pulse slowing down can be scaled down to the domain of millisecond pulses with semiconductor photorefractive crystals.

The coupling of two light beams from a He-Ne laser is studied in undoped $Sn_2P_2S_6$ with a pronounced electron-hole competition in grating formation. The transmission photorefractive gratings are recorded, with the grating vector parallel to the crystallographic **x** axis by two waves polarized also along the **x** axis (Fig. 2).

The intensity of the signal wave is taken to be 3% of the pump wave intensity. The frequency detuning Ω is controlled via a linear (sawtooth) phase shift of the pump wave, introduced with the piezoelectrically driven mirror. The same piezomirror introduces also a low-amplitude (about 0.2 rad) high-frequency cosine phase modulation ($\omega_m \simeq 1.5$ kHz). Such a modulation does not affect the grating recording, but it allows for evaluation of the phase shift in the output signal wave (see, e.g., [11]). This is done by measuring the amplitude of the first harmonic of the modulation frequency ω_m in the transmitted signal with the help of the lock-in amplifier. Direct measurement of the signal wave intensity with and without the pump wave allows for evaluation of intensity coupling in the steady state.

Figure 3 represents the measured dependences of the signal transmission $T = |A_s(\ell)/A_s(0)|^2$ (a),(c) and nonlinear change of propagation constant Δk (b),(d) as a function of frequency detuning, which are typical for nearly compensated dynamic gratings formed by two types of movable charge species. The solid lines in Figs. 3(c) and 3(d) show dependences [9] calculated from

$$\frac{1}{\ell}\ln T = 2\mathrm{Re}\{\gamma\} = 2\gamma_s \frac{1}{1+\Omega^2\tau_s^2} - 2\gamma_f \frac{1}{1+\Omega^2\tau_f^2}, \quad (1)$$

$$\Delta k = \mathrm{Im}\{\gamma\} = -\gamma_s \frac{\Omega\tau_s}{1+\Omega^2\tau_s^2} + \gamma_f \frac{\Omega\tau_f}{1+\Omega^2\tau_f^2}, \quad (2)$$

with the real coupling constants $\gamma_{f,s}$ and decay times $\tau_{f,s}$ for the "fast" and "slow" gratings, respectively. The data of Figs. 3(a) and 3(c) allow one to extract the absolute values for the gain spectrum $\Gamma(\Omega) = 2\mathrm{Re}\{\gamma\}$ and to calibrate the measurements of Δk taking into account Eqs. (1) and (2).

The fast grating which is due to photoexcited holes is shifted in space in this particular experiment in a way to deplete the signal wave. Because of development of the fast grating the attenuation of the signal intensity occurs within a relatively broad spectral range. The width of this resonance of about ± 200 Hz is related to the buildup time of this grating, $\tau_f \simeq 0.8$ ms. When the fast grating is already developed the thermally excited electrons start to move in the nonuniform electric field to compensate it.

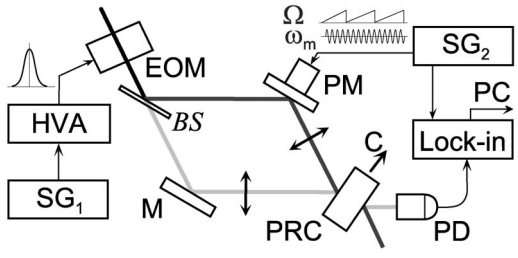

FIG. 2. Schematic of the experimental setup with photorefractive crystal (PRC), electro-optic modulator (EOM), beam splitter (BS), mirror (M), piezoelectrically driven mirror (PM), photodiode (PD), signal generators (SG1 and SG2), high voltage amplifier (HVA), and lock-in amplifier. The arrow near the sample indicates the positive direction of the spontaneous polarization; double arrows show the polarization of the light waves.

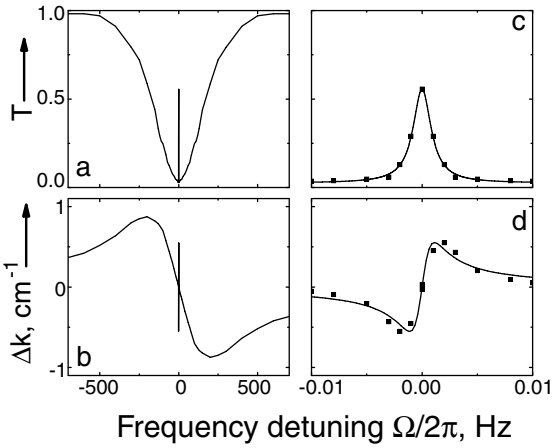

FIG. 3. Experimental dependences of sample transmission T (a),(c) and variation of propagation constant Δk (b),(d) of a weak signal wave on frequency detuning of the signal wave.

The buildup time of the slow electron grating is about $\tau_s \simeq 80$ s and therefore the relevant resonance is much narrower. It can be seen that exactly at degenerate frequency the diffraction "bleaching" occurs and signal transmission increases from a few percent to more than 60% [Figs. 3(a) and 3(c)]. Such a partial bleaching is a consequence of incomplete grating compensation; it may be improved in SPS via heating the sample.

The transmission of the signal wave shown in Fig. 3(a) and 3(c) looks similar to that known for cold gases with EIT [12]: a narrow EIT transmission line appears within the strong absorption of the atomic resonance. The measured spectrum of the phase delay resembles the dispersion profile of EIT, too [13]: There is no phase delay for unshifted frequencies, but there are two regions of nearly linear phase variations with the derivatives $dk/d\Omega$ different in sign and quite different in absolute values [Figs. 3(b) and 3(d)].

From the measured data the expected group velocity $v_{\rm gr} = d\Omega/dk$ can be evaluated for long pulses, i.e., for a well developed slow grating. A rough estimate can be done taking a ratio $\Omega_{\rm max}/\Delta k_{\rm max}$ with $\Omega_{\rm max} = 1/\tau_s$ that ensures the largest value $\Delta k_{\rm max}$, $v_{\rm gr} \simeq 1/\tau_s \Delta k_{\rm max}$. With $\tau_s \simeq 80$ s and $\Delta k_{\rm max} \simeq 0.6$ cm^{-1} we get $v_{\rm gr} \simeq 0.02$ cm/s. Note that because of the strong difference of the decay times of the two gratings, $\tau_s \gg \tau_f$, only the first term on the right-hand side of Eq. (2) is important for evaluation of the $v_{\rm gr}$.

Let us now turn to the experiments on slowing down with gratings we recorded by matched pulses. The electro-optic modulator driven by the signal generator is used to tailor the matched pulses with identical Gaussian temporal envelope and durations from 10^{-1} to 10^2 s from a cw laser output wave (Fig. 2). The intensity of the signal pulse is 1000 times smaller than the intensity of the pump pulse. The piezomirror shown in Fig. 2 is immobile in this experiment. The pulse delay time $\Delta\tau$ and the delayed pulse width w at the $1/e$ level are measured as a function of the input pulse width t_0 [Figs. 4(a) and 4(b)]. Similar to the results obtained with a $BaTiO_3$ crystal in Ref. [3] the delay time $\Delta\tau$ is strongly increasing when t_0 is approaching τ_s, i.e., when the contribution of the slow grating and hence the relevant dispersion becomes important [Fig. 4(a)]. The delay time should saturate for t_0 much longer than τ_s, but with the equipment available we were unable to get reliable results for pulses longer than 100 s: The scatter of measured data becomes too large because of poor long-term stability of the fringe position. The width of the delayed pulse, as distinct from the case of cw pumping described in Ref. [3], follows linearly the input pulse width throughout the whole range of t_0 variation [Fig. 4(b)]. The distortion of the delayed pulse is not severe and the output pulse remains roughly Gaussian.

A delay time of about 5.5 s measured for the 9-mm-thick sample gives a velocity 0.16 cm/s; i.e., the slowing down is weaker (roughly 8 times) than it is estimated from the dispersion. The reasons for this discrepancy are related both to the technical problem of working with very long pulses and to the limitation of the delay time because of the finite duration of the pump pulses. As one can see from Fig. 4(a) the delay time is still not saturated for a pulse duration about 100 s; this means that the ultimate slowing down is not reached, too.

It should be taken into account also that the estimate is done for *steady-state* dynamic gratings with a dispersion which is *not changing* during the whole time of pulse propagation. The quantitative description of the pulse

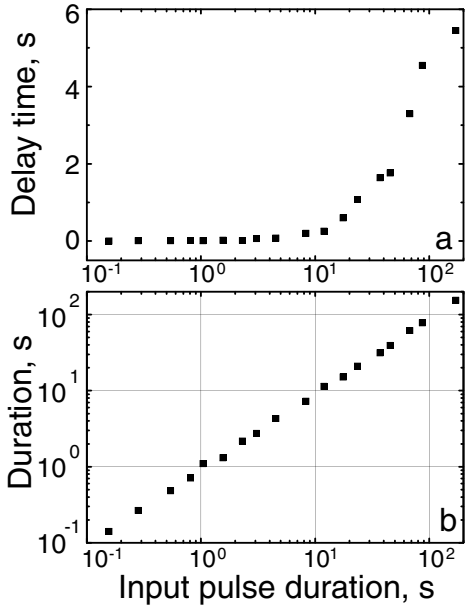

FIG. 4. Time delay of the output pulse maximum (a) and output pulse duration (b) as a function of input pulse duration; $I_p = 1$ W/cm^2, grating spacing $\Lambda = 1.8$ μm.

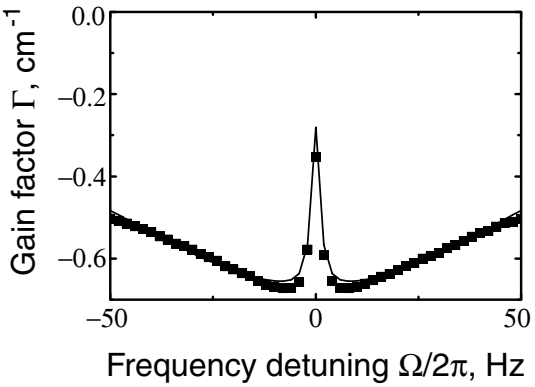

FIG. 5. Two-beam coupling gain spectrum for CdTe:Ge.

propagation should be based, as distinct from the qualitative approach of Ref. [4], on the solution of equations similar to those solved in [3]; the theory [3] should be modified to include the formation of the slow grating.

The slowing down of light pulses in $Sn_2P_2S_6$ has been already discussed in [3]. Shorter pulses have been used in that work, with durations comparable to the decay time of the fast grating, $\tau_f \simeq 1$ ms. The slow grating was not developing within this time range, the compensation did not occur, and the narrow resonance in Fig. 3 did not show up. That is why the slowing down was governed in those experiments by a much more modest dispersion related to the fast grating resonance and the measured pulse velocity was reduced to only 40 cm/s.

The following questions may arise: Is the effect of grating compensation universal and can similar effects of slowing down without gain be demonstrated with faster crystals and consequently with shorter pulses? The answers to both questions are positive. A strong electron-hole competition in space charge formation occurs in many photorefractive crystals [14]. It is known, for example, that the gain factor Γ in CdTe crystals is changing its sign to the opposite when the wavelength of the excitation increases from 0.9 to 1.5 μm [15]. This means that somewhere within this spectral range the amplitude of a grating formed by photoexcited holes becomes equal to that of the grating formed by photoexcited electrons, and complete compensation of two gratings occurs.

We selected particular samples of CdTe:Ge for which such a compensation takes place not far from the Nd^{3+}-doped yttrium aluminum garnet laser wavelength 1.06 μm. Figure 5 represents the gain spectrum $\Gamma(\Omega)$ measured at $\lambda = 1.06$ μm in a similar way as was done for $Sn_2P_2S_6$ at $\lambda = 0.63$ μm. The gain factor in this particular sample is rather modest, but it can go above 1 cm^{-1} in the best CdTe samples with no applied field.

With the decay time of the slow grating $\tau_s \simeq 130$ ms (Fig. 5) the estimated group velocity is about 30 cm/s. This velocity is larger than that reported in this Letter for SPS, and the pulse delay will therefore be smaller; the advantage is, however, in much shorter pulses that can be delayed with CdTe. The other option of working with short pulses is in using higher light intensities: the decay of the photorefractive grating is governed by the dielectric relaxation time which depends on photoconductivity.

To conclude, it is shown in this Letter that giant slowing down of light pulses can be reached in experiments with recording of two out-of-phase dynamic photorefractive gratings using crystals with strong electron-hole competition in space-charge formation. Thus it is proved that photorefractive slowing down can be obtained in media with no gain and no depletion, quite similar to slowing down in EIT experiments.

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