Nearly degenerate two-beam coupling in photorefractive crystals with two species of movable carriers

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Two-beam intensity coupling is calculated for photorefractive crystals with two types of movable charge carrier in the undepleted-pump approximation. The analytical expressions are derived for the temporal evolution of the space-charge field; for weak coupling they are used for calculation of the transmitted beam intensities. The results of the calculation are compared with the experimental observations in photorefractive tin hypothiodiphosphate ($Sn_2P_2S_6$). All experimental data are in reasonable quantitative agreement with the calculations. © 1998 Optical Society of America [S0740-3224(98)03007-0]

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1. INTRODUCTION

The manifestation of the simultaneous presence of two species of movable carrier, electrons and holes, was reported for photorefractive interactions by many authors.[1–4] Nearly always the bipolar transport leads to partial mutual compensation of the space-charge gratings formed by carriers of opposite signs. If the characteristic buildup times for two gratings are quite different the temporal development of the amplified (depleted) wave exhibits typical transient behavior: A relatively fast rise (decrease) of the output intensity is followed by much slower decay (increase) until a steady-state value is reached.

As was shown in Ref. 5, a slight frequency detuning between two recording waves can destroy the grating with slow response time while leaving the amplitude of the fast grating unaffected. The result is considerable improvement of the steady-state gain factor, which may reach the value of the transient peak.

Our purpose in this paper is to calculate the dynamics of two-beam coupling for nearly degenerate in frequency recording waves. The calculations follow the approach of Zhivkova and Miteva.[6] Only the material equations are considered (i.e., the effect of the beam coupling on the contrast of the recording fringes in the bulk of the crystal is neglected). Beam coupling itself is considered to be small enough to permit use of the linear relation between the amplitude of the space-charge grating and the gain factor. Despite the fact that the space-charge limitations are taken into account, the linear recombination of photoexcited carriers is assumed to dominate any multiparticle recombination.

This paper has the following structure. After presenting the initial set of equations (which is exactly the same as in Ref. 6; even the notation is preserved to make the comparison easy) and recalling the main steps of its solution in Section 2, we derive the set of differential equations for the amplitudes of spatially modulated densities of the ionized electron and hole traps with the frequency shift between the writing waves taken into account (Section 3). In Section 4 we solve this set of equations without making any assumptions about the ratio of characteristic grating lifetimes. In Section 5 we consider the particular case of recording in tin hypothiphosphate ($Sn_2P_2S_6$; SPS). Finally, in Section 6 a comparison with the experimental observations is presented.

2. FORMULATION OF THE PROBLEM

A classic arrangement for the recording of the transmission holographic grating in photorefractive crystal is considered. Two waves,

$$E_1 = A_1 \exp(-i\mathbf{k}_1 \cdot \mathbf{r} + i\omega t),$$

$$E_2 = A_2 \exp[-i\mathbf{k}_2 \cdot \mathbf{r} + i(\omega + \Omega)t], \tag{1}$$

form a running fringe pattern

$$I = I_0[1 + m \exp(iKx + i\Omega t)] \tag{2}$$

inside the photorefractive crystal. Here $\mathbf{k}_{1,2}$ are the wave vectors of the recording waves, ω and $(\omega + \Omega)$ are their temporal frequencies, $I_0 = |A_1|^2 + |A_2|^2$ is the total intensity inside the crystal, $m = |2A_1A_2|/I_0$ is the fringe contrast, and K is the spatial frequency of the fringe pattern.

The contrast of the fringes is taken to be small, i.e., $m \ll 1$. The self-diffraction from the recorded grating is

neglected; i.e., m is assumed to be independent of propagation coordinate z. Therefore only the material equations are considered.

Following the approach given in Ref. 6, we assume the presence of two types of impurity center, one being close to the conduction band with total density N_1 and the other close to the valence band with total density N_2. The first level is populated by electrons, and the second is populated by holes. Both levels are partially ionized; ${N_1}^i$ and ${N_2}^i$ are the densities of the empty electron and hole levels. The electrons from level 1 can be excited to the conduction band either thermally (with the probability β_n) or because of phototransition (with probability $s_n I$). Similarly, holes can be released from level 2 to the valence band by thermal transition (probability β_p) or by phototransition (probability $s_p I$).

The rate equations for ionized electron traps and hole traps are

$$\frac{\partial {N_1}^i}{\partial t} = (\beta_n + s_n I)(N_1 - {N_1}^i) - \gamma_n {N_1}^i n,$$

$$\frac{\partial {N_2}^i}{\partial t} = -(\beta_p + s_p I){N_2}^i + \gamma_p (N_2 - {N_2}^i)p, \qquad (3)$$

where n and p are the densities and $\gamma_{n,p}$ are the recombination constants for free electrons and free holes, respectively.

The continuity equations for electrons and holes are

$$\frac{\partial n}{\partial t} = \frac{\partial {N_1}^i}{\partial t} + \frac{1}{e}\frac{\partial j_n}{\partial x},$$

$$\frac{\partial p}{\partial t} = -\frac{\partial {N_2}^i}{\partial t} - \frac{1}{e}\frac{\partial j_p}{\partial x}, \qquad (4)$$

and the current equations are

$$j_n = e\mu_n nE + eD_n \frac{\partial n}{\partial x},$$

$$j_p = e\mu_p pE - eD_p \frac{\partial p}{\partial x}, \qquad (5)$$

where $\mu_{n,p}$ are the mobilities, $D_{n,p} = (k_B T/e)\mu_{n,p}$ are the diffusivities for electrons and holes, respectively, and $j_{n,p}$ are the electron and hole currents.

The Poisson equation completes the set of material equations for calculation of the space-charge field:

$$\frac{\partial E}{\partial x} = \frac{e}{\epsilon\epsilon_0}(p - n + {N_1}^i - {N_2}^i - N_a), \qquad (6)$$

where $\epsilon\epsilon_0$ is the dielectric constant and N_a is the density of optically inactive acceptors that are compensating for ${N_1}^i$ and ${N_2}^i$.

3. EQUATIONS FOR THE SPACE CHARGE

The solution of the set of material equations [Eqs. (3)–(6)] will be found for low contrast [$m \ll 1$ in Eq. (2)] when we can represent all spatially dependent quantities, keeping only the first nonvanishing spatially nonuniform terms in expansions, as

$$n(t, x) = n_0(t) + n_1(t)\exp(iKx + i\Omega t) + \text{c.c.},$$

$$p(t, x) = p_0(t) + p_1(t)\exp(iKx + i\Omega t) + \text{c.c.},$$

$${N_1}^i(t, x) = \bar{N}_1(t) + M_1(t)\exp(iKx + i\Omega t) + \text{c.c.},$$

$${N_2}^i(t, x) = \bar{N}_2(t) + M_2(t)\exp(iKx + i\Omega t) + \text{c.c.},$$

$$E(t, x) = E_1(t)\exp(iKx + i\Omega t) + \text{c.c.} \qquad (7)$$

Here $E_1(t)$ is the amplitude of the space-charge field.

By substituting Eqs. (7) into Eqs. (4) and combining the terms that describe the spatially uniform quantities and terms with identical exponential functions we get for adiabatically slow temporal variations ($\partial n/\partial t = \partial p/\partial t = 0$)

$$\bar{N}_1 = \text{const.},$$

$$n_0 = (\beta_n + s_n I_0)(N_1 - \bar{N}_1)/\gamma_n \bar{N}_1,$$

$$\bar{N}_2 = \text{const.},$$

$$p_0 = (\beta_p + s_p I_0)\bar{N}_2/\gamma_p (N_2 - \bar{N}_2) \qquad (8)$$

and for the amplitudes of the spatially modulated densities

$$n_1 = \left[m s_n I_0 (N_1 - \bar{N}_1) - M_1(\beta_n + s_n I_0 + \gamma_n n_0 + i\Omega) - \frac{\partial M_1}{\partial t} \right] \Big/ \gamma_n \bar{N}_1,$$

$$p_1 = \left[m s_p I_0 \bar{N}_2 - M_2(\beta_p + s_p I_0 + \gamma_p p_0 + i\Omega) - \frac{\partial M_2}{\partial t} \right] \Big/ \gamma_p (N_2 - \bar{N}_2). \qquad (9)$$

The substitution of Eqs. (7) into Poisson equation (6) will provide

$$N_a = p_0 - n_0 + \bar{N}_1 - \bar{N}_2 \qquad (10)$$

and for the space-charge field

$$E_1 = (p_1 - n_1 + M_1 - M_2)c/(\epsilon\epsilon_0 iK). \qquad (11)$$

By a similar procedure with Eqs. (4) and (5) we get

$$\frac{\partial M_1}{\partial t} + i\Omega M_1 + \frac{e}{\epsilon\epsilon_0}\mu_n n_0 (p_1 - n_1 + M_1 - M_2) - K^2 D_n n_1 = 0,$$

$$\frac{\partial M_2}{\partial t} + i\Omega M_2 - \frac{e}{\epsilon\epsilon_0}\mu_p p_0 (p_1 - n_1 + M_1 - M_2) - K^2 D_p p_1 = 0. \qquad (12)$$

Introducing the standard characteristic times

$${\tau_m}^n = \frac{\epsilon\epsilon_0}{e\mu_n n_0}$$

(dielectric relaxation time for electrons in the dark),

$${\tau_m}^p = \frac{\epsilon\epsilon_0}{e\mu_p p_0}$$

(dielectric relaxation time for holes in the dark),

$$\tau_n = \frac{1}{\gamma_n \bar{N}_1} \quad \text{(free-electron lifetime)},$$

$$\tau_p = \frac{1}{\gamma_p (N_2 - \bar{N}_2)} \quad \text{(free-hole lifetime)}, \tag{13}$$

the characteristic transport lengths

$$L_{Dn}{}^2 = D_n \tau_n \quad \text{(electron diffusion length)},$$

$$L_{Dp}{}^2 = D_p \tau_p \quad \text{(hole diffusion length)}, \tag{14}$$

and the characteristic electric fields

$$E_{qn} = \frac{e\bar{N}_1(N_1 - \bar{N}_1)}{\epsilon\epsilon_0 K N_1} \quad \text{(limiting space-charge field that could be created by the electrons)},$$

$$E_{qp} = \frac{e\bar{N}_2(N_2 - \bar{N}_2)}{\epsilon\epsilon_0 K N_2} \quad \text{(limiting space-charge field that could be created by the holes)}, \tag{15}$$

and combining Eqs. (14) and (15) with Eqs. (10) and (11), we complete the set of linear differential equations for the amplitudes of space-modulated densities for ionized electron traps M_1 and ionized hole traps M_2:

$$\begin{aligned}
&\frac{\partial M_1}{\partial t}\left(1 + \frac{\tau_n}{\tau_m{}^n} + K^2 D_n \tau_n\right) \\
&\quad + M_1\left[\frac{1 - \tau_n(\beta_n + s_n I_0 + \gamma_n n_0 + i\Omega)}{\tau_m{}^n}\right. \\
&\quad \left. + K^2 D_n \tau_n(\beta_n + s_n I_0 + \gamma_n n_0 + i\Omega)\right] + \frac{\partial M_2}{\partial t}\left(-\frac{\tau_p}{\tau_m{}^p}\right) \\
&\quad + M_2\left[-\frac{1 + \tau_p(\beta_p + s_p I_0 + \gamma_p p_0 + i\Omega)}{\tau_m{}^p}\right] \\
&\qquad = -\frac{\tau_p}{\tau_m{}^n} m s_p I_0 \bar{N}_2 + \frac{\tau_n}{\tau_m{}^n} m s_n I_0 (N_1 - \bar{N}_1) \\
&\qquad\quad + K^2 D_n \tau_n m s_n I_0 (N_1 - \bar{N}_1), \\
&\frac{\partial M_2}{\partial t}\left(1 + \frac{\tau_p}{\tau_m{}^p} + K^2 D_p \tau_p\right) \\
&\quad + M_2\left[\frac{1 - \tau_p(\beta_p + s_p I_0 + \gamma_p p_0 + i\Omega)}{\tau_m{}^p}\right. \\
&\quad \left. + K^2 D_p \tau_p(\beta_p + s_p I_0 + \gamma_p p_0 + i\Omega)\right] + \frac{\partial M_1}{\partial t}\left(-\frac{\tau_n}{\tau_m{}^n}\right) \\
&\quad + M_1\left[-\frac{1 + \tau_n(\beta_n + s_n I_0 + \gamma_n n_0 + i\Omega)}{\tau_m{}^n}\right] \\
&\qquad = -\frac{\tau_n}{\tau_m{}^p} m s_n I_0 (N_1 - \bar{N}_1) + \frac{\tau_p}{\tau_m{}^p} m s_p I_0 \bar{N}_2 \\
&\qquad\quad + K^2 D_p \tau_p m s_p I_0 \bar{N}_2 .
\end{aligned} \tag{16}$$

Depending on the experimental conditions, Eqs. (16) can be considerably simplified. It is well known, e.g., that the free-carrier lifetime is much smaller than the dielectric relaxation time. Therefore the terms that contain $(\tau_p/\tau_m{}^n)$, $(\tau_n/\tau_m{}^n)$, $(\tau_p/\tau_m{}^p)$, and $(\tau_n/\tau_m{}^p)$ can be neglected; then Eqs. (16) will be reduced to the form

$$\begin{aligned}
&\frac{\partial M_1}{\partial t}(1 + K^2 D_n \tau_n) + \frac{M_1}{\tau_m{}^n}[1 + i\Omega\tau_m{}^n \\
&\quad + K^2 D_n \tau_n \tau_m{}^n(\beta_n + s_n I_0 + \gamma_n n_0 + i\Omega)] + M_2\left(-\frac{1}{\tau_m{}^n}\right) \\
&\qquad = K^2 D_n \tau_n m s_n I_0 (N_1 - \bar{N}_1),
\end{aligned}$$

$$\begin{aligned}
&\frac{\partial M_2}{\partial t}(1 + K^2 D_p \tau_p) + \frac{M_2}{\tau_m{}^p}[1 + i\Omega\tau_m{}^p \\
&\quad + K^2 D_p \tau_p \tau_m{}^p(\beta_p + s_p I_0 + \gamma_p p_0 + i\Omega)] + M_1\left(-\frac{1}{\tau_m{}^p}\right) \\
&\qquad = K^2 D_p \tau_p m s_p I_0 \bar{N}_2 ,
\end{aligned} \tag{17}$$

with

$$\begin{aligned}
A_1 &= -B_1[1 + i\Omega\tau_m{}^n(1 + l_{Dn}{}^2 K^2) + (E_D/E_{qn})], \\
A_2 &= -[\tau_m{}^p(1 + L_{Dp}{}^2 K^2)]^{-1}, \\
B_1 &= -[\tau_m{}^n(1 + L_{Dn}{}^2 K^2)]^{-1}, \\
B_2 &= -A_2[1 + i\Omega\tau_m{}^p(1 + l_{Dp}{}^2 K^2) + (E_D/E_{qp})], \\
C_1 &= -(m\epsilon\epsilon_0/e) K E_D B_1 \frac{s_n I_0}{\beta_n + s_n I_0}, \\
C_2 &= -(m\epsilon\epsilon_0/e) K E_D A_2 \frac{s_p I_0}{\beta_p + s_p I_0},
\end{aligned} \tag{18}$$

and the set of equations for $M_{1,2}$ becomes

$$\begin{aligned}
\frac{\partial M_1}{\partial t} + A_1 M_1 + B_1 M_2 &= C_1, \\
\frac{\partial M_2}{\partial t} + A_2 M_1 + B_2 M_2 &= C_2 .
\end{aligned} \tag{19}$$

With zeroth initial conditions (no gratings at $t = 0$) the solutions of Eqs. (17) are

$$\begin{aligned}
M_1(t) &= -\frac{B_1 C_2 - B_2 C_1}{\alpha_1 \alpha_2} - \frac{\alpha_1 + B_2}{A_2} K_1 \exp(\alpha_1 t) \\
&\quad - \frac{\alpha_2 + B_2}{A_2} K_2 \exp(\alpha_2 t), \\
M_2(t) &= -\frac{A_2 C_1 - A_1 C_1}{\alpha_1 \alpha_2} + K_1 \exp(\alpha_1 t) \\
&\quad + K_2 \exp(\alpha_2 t).
\end{aligned} \tag{20}$$

Taking into account that the densities of free electrons and free holes are usually much smaller than the densities of the charge redistributed in traps, i.e., p_1, $n_1 \ll M_1$, M_2, we get from Eqs. (13) for the space-charge field

$$E_{sp} = \frac{ie}{\epsilon\epsilon_0 K}(M_2 - M_1), \tag{21}$$

where

$$K_1 = \frac{\alpha_1 C_2 + A_1 C_2 - A_2 C_1}{\alpha_1(\alpha_1 - \alpha_2)},$$

$$K_2 = -\frac{\alpha_1 C_2 + A_1 C_2 - A_2 C_1}{\alpha_2(\alpha_1 - \alpha_2)}; \tag{22}$$

$$\alpha_{1,2} = \frac{-(A_1 + B_2) \pm [(A_1 - B_1)^2 + 4A_2B_1]^{1/2}}{2}. \tag{23}$$

This is a general solution for the space charge for an arbitrary ratio of relaxation times of two gratings and arbitrary frequency detuning. It is easy to verify that for $\Omega = 0$ this solution coincides with that obtained by Zhivkova and Miteva.[6]

As the refractive-index change is proportional to the space-charge field in the approximation of the theory that we are considering, Eq. (21) also describes the dynamics of the phase grating amplitude. For $\Omega = 0$ this grating is $\pi/2$ shifted with respect to the fringes, and the coupling strength is purely real. With $\Omega \neq 0$ the phase shift deviates from the exact $\pi/2$ value, and therefore the coupling strength becomes complex. To calculate the gain factor that is usually measured in the experiment one should take the real part of the complex coupling constant:

$$\Gamma = \mathrm{Re}[(2\pi i n^3 r_{\mathrm{eff}} E_{sc})/m\lambda], \tag{24}$$

where r_{eff} is the effective electro-optic constant and E_{sc} is given by Eqs. (22).

In Section 4 we shall concentrate on the interaction with the parameters that is typical of SPS and present a detailed comparison of calculated data with those measured in our previous experiment.[5]

4. COMPARISON WITH EXPERIMENTAL RESULTS FOR TIN HYPOTHIODIPHOSPHATE

According to the data of Ref. 5, the formation of one of two out-of-phase gratings in SPS is due to photostimulated charge separation, whereas for the other grating the thermally stimulated charge transport is dominating. Figure 1 is the energy-level diagram of SPS. This result allows us further to simplify the model by putting $s_p = 0$ and therefore $C_2 = 0$. Taking into account the large difference in characteristic relaxation times for the two gratings ($\simeq$17 ms for the fast grating and more than 100 s for the slow grating), we can present the solution for $M_{1,2}$ in the following form:

$$M_1 \simeq [1 - \exp(-A_1 t)]C_1/A_1,$$

$$M_2 \simeq -[1 - \exp(-B_2 t)]C_1A_2/A_1B_2. \tag{25}$$

Note the absence of the fast grating lifetime in the first of Eqs. (25). The equation looks reasonable, however, as the time needed to form the fast grating is much less than time necessary to develop the slow grating.

The ultimate expression for the gain factor is

$$\Gamma(t) \propto \frac{E_D/(1 + l_{Sn}{}^2K^2)}{1 + (\tau_m{}^n\Omega)^2(1 + l_{Dn}{}^2K^2)^2/(1 + l_{Sn}{}^2K^2)^2} \times \left(-\frac{1 + l_{Sp}{}^2K^2}{(1 + l_{Sp}{}^2K^2)^2 + (\tau_m{}^p\Omega)^2(1 + l_{Dp}{}^2K^2)^2} \times \left\{1 - \left[\frac{(\tau_m{}^p\Omega)(1 + l_{Dp}{}^2K^2)}{1 + l_{Sp}{}^2K^2}\sin(\Omega t) + \cos(\Omega t)\right]\exp\left(-\frac{t}{\tau_m{}^p}\frac{1 + l_{Sp}{}^2K^2}{1 + l_{Dp}{}^2K^2}\right)\right\} + \left\{1 - \left[\frac{(\tau_m{}^n\Omega)(1 + l_{Dn}{}^2K^2)}{1 + l_{Sn}{}^2K^2}\sin(\Omega t) + \cos(\Omega t)\right]\exp\left(-\frac{t}{\tau_m{}^n}\frac{1 + l_{Sn}{}^2K^2}{1 + l_{Dn}{}^2K^2}\right)\right\}\right). \tag{26}$$

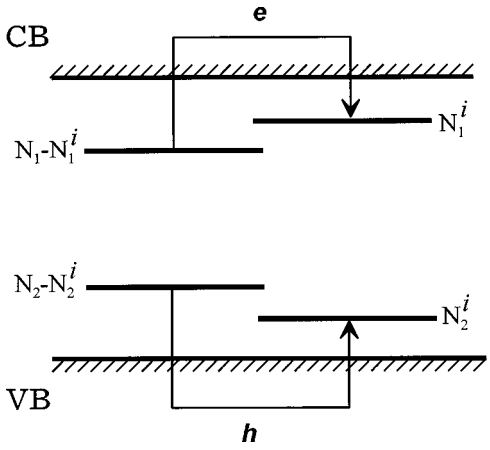

Fig. 1. Energy-level diagram. CB, conduction band; VB, valence band.

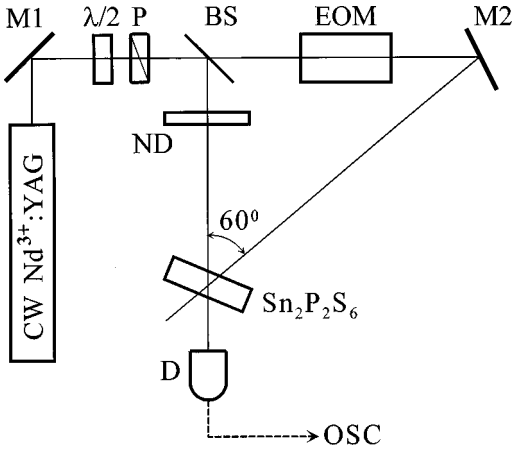

Fig. 2. Schematic representation of the experimental setup. M's, mirrors; P, prism; BS, beam splitter; EOM, electro-optic modulator; ND, neutral-density filter; D, detector; OSC, oscillator.

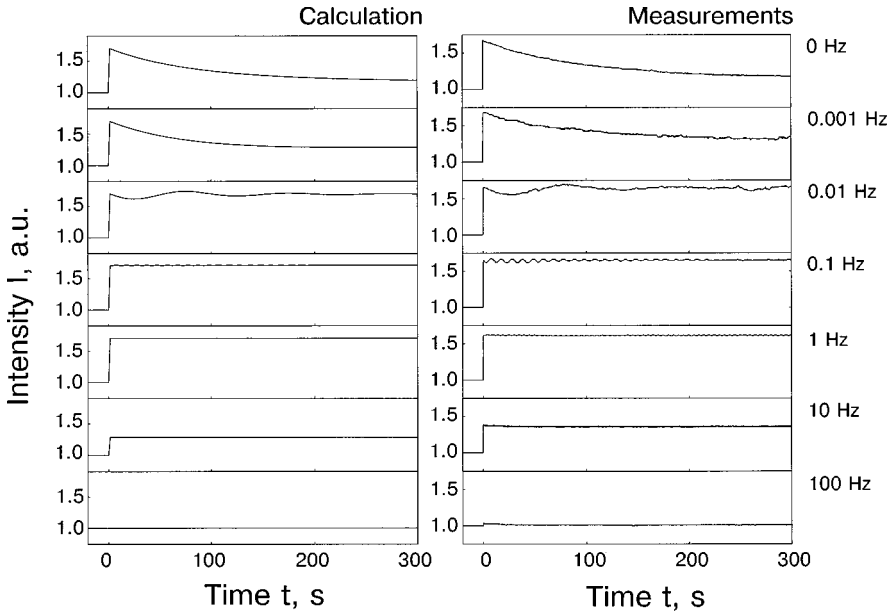

Fig. 3. Calculated (left) and measured (right) temporal variations of the transmitted signal wave intensity. The total intensity of the two light waves is 15 W/cm^2 at $\lambda = 1\ \mu$m; the fringe spacing $\Lambda = 1\ \mu$m.

Table 1. Parameters of the SPS Sample Used in the Calculation

τ_m^p (s)	τ_m^n (s)	l_{Sn} (μm)	l_{Sp} (μm)	$\frac{4\pi^2 n^3 r_{\text{eff}}}{\lambda \cos\theta'} \frac{k_B T}{e}$
140	0.03	0.9	0.65	7.1×10^{-4}

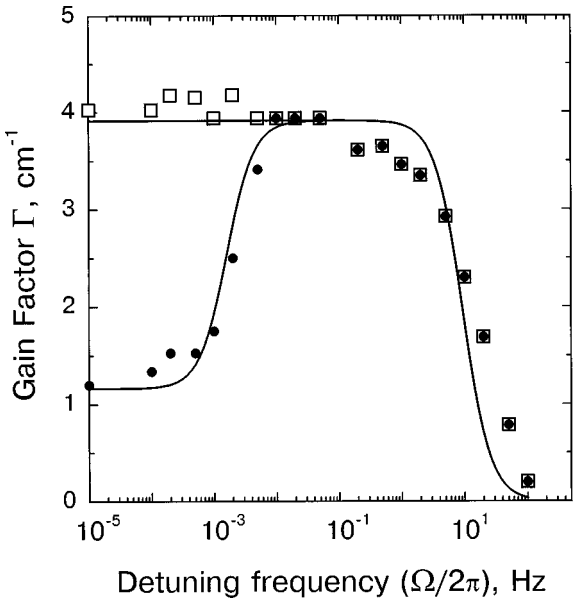

Fig. 4. Transient (open squares) and steady-state (filled circles) gain factor versus frequency detuning. The solid curve shows the results calculated with the parameters given in Table 1.

Relation (26) describes the dynamics of the gain factor in SPS for two waves shifted by Ω in frequency with respect to each other. Let us first discuss some limiting cases and verify that relation (26) can be reduced to the known results for specific experimental situations.

A. Two Gratings, Strictly Degenerate Interaction

$$\Gamma(t) \propto \frac{E_D}{1 + {l_{Sn}}^2 K^2} \left\{ -\frac{1}{(1 + {l_{Sp}}^2 K^2)} \times \left[1 - \exp\left(-\frac{t}{{\tau_m}^p} \frac{1 + {l_{Sp}}^2 K^2}{1 + {l_{Dp}}^2 K^2} \right) \right] + \left[1 - \exp\left(-\frac{t}{{\tau_m}^n} \frac{1 + {l_{Sn}}^2 K^2}{1 + {l_{Dn}}^2 K^2} \right) \right] \right\}.$$

This relation describes the characteristic dynamics of the beam coupling with a transient peak followed by a much smaller steady-state gain.[6–8]

B. Two Gratings, Strictly Degenerate Interaction, Saturation

$$\Gamma(t \to \infty) \propto \frac{E_D {l_{Sp}}^2 K^2}{(1 + {l_{Sn}}^2 K^2)(1 + {l_{Sp}}^2 K^2)}.$$

An important consequence of this equation is that for small spatial frequencies the steady-state gain factor increases as K^3 with a constant of proportionality that includes the diffusion length for holes ${l_{Sp}}^2$.

C. Only One (Fast) Grating

To suppress the slow grating let us assume that its Debye screening length is infinitely large. In this case

$$\Gamma(t) \propto \frac{E_D(1 + {l_{Sn}}^2 K^2)}{(1 + {l_{Sn}}^2 K^2)^2 + ({\tau_m}^n \Omega)^2 (1 + {l_{Dn}}^2 K^2)^2} \times \left\{ 1 - \left[{\tau_m}^n \Omega \frac{1 + {l_{Dn}}^2 K^2}{1 + {l_{Sn}}^2 K^2} \sin(\Omega t) + \cos(\Omega t) \right] \times \exp\left(-\frac{t}{{\tau_m}^n} \frac{1 + {l_{Sn}}^2 K^2}{1 + {l_{Dn}}^2 K^2} \right) \right\}.$$

D. Only One Grating, Saturation

The equation

$$\Gamma(t \to \infty) \propto \frac{E_D}{(1 + l_{Sn}{}^2K^2)[1 + (\tau_m{}^n\Omega)^2(1 + l_{Dn}{}^2K^2)^2/(1 + l_{Sn}{}^2K^2)^2]}$$

describes the well-known Lorentzian profile of the gain spectrum for nearly degenerate two-beam coupling in a medium with single exponential decay.[9,10] One can also obtain it by taking the Fourier transform of the characteristic grating decay curve. Note that HWHM of the Lorentz profile depends on the real grating lifetime, i.e., on the dielectric relaxation time with the correction factor including transport lengths l_{Dn} and l_{Sn}.

E. Only One Grating, Saturation, Degenerate Interaction

$$\Gamma(t \to \infty) \propto \frac{E_D}{(1 + l_{Sn}{}^2K^2)}.$$

As expected, the gain factor depends on the diffusion field with the correction factor that is due to the screening effect.[11]

We now return to the analysis of qualitative consequences of relation (26):

1. With increasing frequency detuning Ω the contribution of the slow grating gradually decreases; see the factor

$$\frac{1 + l_{Sp}{}^2K^2}{(1 + l_{Sp}{}^2K^2)^2 + (\tau_m{}^p\Omega)^2(1 + l_{Dp}{}^2K^2)^2}$$

in the left-hand side of relation (26), with Ω^2 in the denominator.

2. For the frequency detuning range $(1/\tau_m{}^p) \ll \Omega \ll (1/\tau_m{}^n)$ the steady-state (saturated) gain factor is nearly equal to the gain factor that is due to the fast grating alone (the main contribution in the gain factor comes from the term responsible for the fast grating):

$$\Gamma(t) \propto \frac{E_D/(1 + l_{Sn}{}^2K^2)}{1 + (\tau_m{}^n\Omega)^2(1 + l_{Dn}{}^2K^2)^2/(1 + l_{Sn}{}^2K^2)^2} \times \left[1 - \cos(\Omega t)\exp\left(-\frac{t}{\tau_m{}^n}\frac{1 + l_{Sn}{}^2K^2}{1 + l_{Dn}{}^2K^2}\right)\right].$$

3. For nearly degenerate interaction the dynamics exhibit damped oscillations; the larger Ω is, the smaller the amplitudes of the oscillations become.

4. The frequency of the damped oscillations is exactly equal to the frequency detuning Ω.

Qualitatively all these features were observed in the experiments with SPS.[5] Figure 2 represents the experimental setup for the investigation of beam coupling. We use an electro-optic modulator for frequency detuning. In Fig. 3 we show the measured dependence of the amplified signal intensity along with that calculated for the experimental parameters given in Table 1. Except for the Debye screening length l_{Sp}, which is taken from Ref. 8 (sample 2), the data in this table are from Ref. 5.

One can see excellent agreement between the calculated and the measured data. In Fig. 4 we plot the calculated dependences of the steady-state and transient gain factors versus detuning frequency and compare them with the experimental dependences taken from our publication.[5] Once again we note the reasonable agreement between the experimental and the calculated data.

5. CONCLUSIONS

In conclusion, we have presented a theory that permits us to calculate the dynamics of space-charge grating formation and decay for the case of bipolar conductivity. The general solution was derived for relatively weak interaction (or small interaction length) for cases in which beam coupling effects can be neglected and in which the contrast of the recording fringes can be considered to be independent of the propagation coordinate.

We analyzed in detail the particular case of $Sn_2P_2S_6$, for which one grating is formed by the light-induced redistribution of charges and the second grating, compensating in part for the first grating, appears as a consequence of thermal redistribution of the charge carriers of opposite sign. Despite the fact that charge hopping[8] is believed to be the main process of charge transport in $Sn_2P_2S_6$, the comparison with the theory developed for the band transport gives reasonable quantitative agreement of the measured and the calculated data. This result demonstrates again that from purely wave mixing experiments it is not easy to distinguish between band transport and charge hopping transport in grating formation.

We believe that this calculation can be successfully used for other photorefractive crystals that exhibit bipolar conductivity.

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