Modeling of the photorefractive nonlinear response in $Sn_2P_2S_6$ crystals

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We develop a theory of the photorefractive nonlinear response for $Sn_2P_2S_6$ crystals. The theory incorporates two types of charge carrier (optically active and passive), provides explicit expressions for the characteristic buildup–relaxation rates and gain factors, explains naturally a big variety of accumulated experimental data, and facilitates characterization–optimization of this important nonlinear material. © 2007 Optical Society of America

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1. INTRODUCTION

Ferroelectric crystals of $Sn_2P_2S_6$ (SPS) are recognized nowadays as an important material for photorefractive (PR) applications, especially in the red and near-infrared spectral ranges.[1] The attractive features of this new PR material are fast response, high sensitivity, and strong nonlinearity.

The presence of two characteristic times [fast (intensity dependent) and slow (intensity independent)] is typical for the PR response in SPS crystals, at least in nominally undoped (yellow) ones.[1–3] It is attributed to two types of charge carrier, one of which is photoexcited (active) and the other is passive, i.e., is not directly affected by light. Several important features of the PR response in SPS crystals, such as the compensation character of recording and beam-coupling kinetics[1,3] and a narrow dip in the frequency response,[4,5] are also due to the presence of the charge compensation.

The basic model with two types of charge carrier for description of the PR response is well known in the literature, see, e.g., Ref. 6 and references therein. It incorporates the collective character of the charge motion, includes many variable internal parameters, and can, depending on the choice of these parameters, be applied to different materials and experimental situations.

The first attempt to apply the two-carrier model to SPS crystals was made in Ref. 5 in 1998. The compensation character of the PR recording and the frequency dependence of the PR response were described; this allowed researchers to explain later the behavior of the SPS-based ring-loop PR oscillator.[7] Much less attention in Ref. 5 was paid to establish the interrelations among the observable fast and slow response times, the individual properties of the two systems of charge carrier (the effective tap concentration, the dielectric relaxation times, etc.), and the experimental parameters, such as the grating period. This important aspect of the SPS modeling was missed despite the fact that the initial general expression for the characteristic response times was correctly and properly derived. Moreover, the general solution for the grating amplitudes was not properly adapted for the SPS parameters, and the slow response time was misinterpreted as the dielectric relaxation time of the passive charge carriers.

Our concern is that this interpretation of the slow response time is not consistent with the assumption about the presence of two types of charge carrier. The relaxation rates in this case are expected to be the total rate of dielectric relaxation (caused by the total conductivity) and the rate of ambipolar diffusion.[8] The first of them gives the reciprocal fast response time; it is close to the dielectric relaxation time for the photoexcited charge carriers in the case of SPS. The second relaxation rate gives the reciprocal slow response time; it should depend strongly on the grating period and, typically, be much longer than the dielectric relaxation time for the passive charge carriers.

Furthermore, it turns out that the situation with the PR response of SPS crystals is similar to the situation with the thermal fixing phenomenon in $LiNbO_3$ crystals, which also incorporates photoexcited and passive (compensating) charge carriers.[9] The theory of fixing, which employs self-consistently the collective motion of the charge carriers, is well developed.[10]

The purpose of this paper is to formulate a self-

consistent theory of the PR response in SPS crystals, to interpret the main experimental facts within this theory, and to provide relations for evaluation of the crystal parameters. The difference from the theory of thermal fixing in $LiNbO_3$ crystals comes mainly from different ranges of the actual material and experimental parameters. The main ingredient of the theory is material relations that enable one to express the space-charge field and nonlinear index change by the light amplitudes. Being supplemented by the standard equations for the light amplitudes (describing Bragg diffraction), these relations allow one to analyze any optical PR phenomenon in SPS crystals.

We will see below that a part of the previous relations for the PR response of SPS crystals (namely, the relations describing the initial fast stage of the PR recording) holds true and can be used for quantitative comparison with experiment. At the same time, the part of the relations that is relevant to the slow component, to the steady state, and to the frequency response experiences substantial quantitative (but not qualitative) changes. Our modified theory gives additional possibilities for interpretation of the accumulated experimental data and for determination of material parameters. The theory predicts also additional features of the PR response in SPS crystals; they are related mainly to the dependence of the observable characteristics on the grating period. Note, lastly, that the experimentally detected strong dependence of the slow response time on the grating period[3,7] fits well our theory and does not fit the suggestion of Ref. 5 about this time.

The text is structured as follows. First, we formulate shortly the basic model and specify the assumptions made. Then we derive coupled dynamical equations for the amplitudes of space-charge gratings and determine the characteristic fast and slow relaxation times. Next, we describe the two-step (fast–slow) recording kinetics of the space-charge field and the dependence of the PR response on the frequency detuning between the incident light waves. In the end, we discuss the relationship between theory and experiment and draw conclusions.

2. BASIC MODEL

The PR nonlinear index change Δn is due to the light-induced space-charge field E_{sc} and the linear electro-optic effect.[6] It is traditionally represented by $\Delta n=-n^3 r E_{sc}/2$, where n is the background refractive index and r is the relevant electro-optic coefficient. The PR nonlinear response can thus be described by relations expressing E_{sc} through the recording light amplitudes. These relations essentially depend on the charge-transport mechanism in question.

In accordance with Refs. 1, 3, and 11, we assume that there are two types of mobile charge carrier. The carriers of the first type are negative and optically passive; their concentration is H, and the averaged (in z) concentration is H_0. The carriers of the second type are positive and optically excited; their concentration is p, and the averaged concentration is p_0. The concentration of traps filled with electrons is N, the corresponding averaged concentration is N_0, the total trap concentration is N_Σ, and the concentration of compensating charges (responsible for neutrality of the crystal) is denoted N_C. Then the set of coupled equations for E_{sc}, N, H, and p can be presented in the following standard form:

$$\frac{\partial E_{sc}}{\partial z}=-\frac{e}{\epsilon\epsilon_0}(N-N_C+H-H_0-p),$$

$$\frac{\partial N}{\partial t}=s_i I(N_\Sigma-N)-s_r N_p,$$

$$\frac{\partial p}{\partial t}=\frac{\partial N}{\partial t}-\frac{1}{e}\frac{\partial j_p}{\partial z},$$

$$\frac{\partial H}{\partial t}=\frac{1}{e}\frac{\partial j_H}{\partial z}. \tag{1}$$

Here s_i and s_r are the ionization and recombination constants, I is the light intensity, and j_p and j_H are the partial current densities,

$$j_p=e\mu_p p E_{sc}-eD_p\frac{\partial p}{\partial z},$$

$$j_H=e\mu_H H E_{sc}+eD_H\frac{\partial H}{\partial z}, \tag{2}$$

where e is the charge quantum, μ_p and μ_H are the mobilities of the photoexcited and passive charges, $D_p=\mu_p k_B T/e$ and $D_H=\mu_H k_B T/e$ are their diffusion coefficients, k_B is the Boltzmann constant, and T is the absolute temperature. It is assumed that (i) all variables depend only on the time t and the spatial coordinate z and (ii) no external electric field is applied to the crystal so that the only mechanism of creation of the space-charge field is diffusion. The latter assumption is fulfilled for all experimental publications on SPS known to date, except Ref. 12.

To describe the PR response, we suppose that the light intensity I is modulated inside the crystal via interference of two frequency-degenerate (or almost degenerate) plane monochromatic waves with complex amplitudes A_1 and A_2,

$$I=I_0\left(1+\frac{m}{2}e^{iKz}+\frac{m^*}{2}e^{-iKz}\right), \tag{3}$$

where z is the fringe coordinate, $I_0=|A_1|^2+|A_2|^2$ is the average (in z) intensity, K is the spatial frequency (the absolute value of the grating vector), $m=2A_1A_2^*/I_0$ is the complex modulation coefficient (its absolute value is the fringe contrast), and the asterisk stands for complex conjugation. In the general case, a frequency detuning Ω exists between the light waves; i.e., the light pattern is moving along z and $m\propto\exp(-i\Omega t)$.

The following conventional assumptions are expected to be fulfilled:

—The usual adiabatic approximation for photoexcited carriers. This means that the lifetime of photoexcited charge carriers $\tau_p=(s_r N_C)^{-1}$ is much shorter than the

characteristic relaxation times of the space-charge field. It is typically fulfilled with a large margin of safety in the cw intensity range.

—The linear contrast approximation. It is the conventional approximation for the description of the PR response. In ferroelectrics it is valid usually up to the values of light contrast $|m| \approx 1$.

—The low-intensity approximation implying that the concentration of photoexcited carriers is much smaller than that of the trapped ones. It is not specific for the case in question.

Although the above assumptions are not different from the assumptions of the thermal fixing theory,[10] some important differences from the case of $LiNbO_3$ crystals have to be mentioned: the photovoltaic charge transport is negligible in SPS; the diffusion and Debye screening lengths are not expected to be small compared with K^{-1}; the temperature T is not expected to be high; and the characteristic relaxation times are much shorter in SPS, which shifts the accent to the studies of the slow processes.

Formally, the passive mobile charges are ions according to Eqs. (1) and (2). We have made sure, however, that all subsequent results for the observable characteristics remain the same under the assumption that the compensating charges are thermally excited electrons. Our theory is thus fairly general concerning the nature of the compensating charges.

3. DYNAMICAL EQUATIONS

Within the considered model, the space-charge field E_{sc}, the variation of the electron density on the traps $N-N_0$, and the variation of the passive (compensating) charge density $H-H_0$ can be represented as

$$E_{sc} = E_K e^{iKz} + \text{c.c.},$$

$$N - N_0 = N_K e^{iKz} + \text{c.c.},$$

$$H - H_0 = H_K e^{iKz} + \text{c.c.}, \tag{4}$$

where E_K,N_K, and H_K are the corresponding amplitudes and c.c. means complex conjugate. Similarly, one can introduce the index grating amplitude Δn_K, which is proportional to the field amplitude E_K.

The amplitudes N_K and H_K obey the set of coupled equations[10]

$$\frac{dN_K}{dt} + \gamma_{11} N_K + \gamma_{12} H_K = F_K,$$

$$\frac{dH_K}{dt} + \gamma_{21} N_K + \gamma_{22} H_K = 0. \tag{5}$$

In accordance with the physical situation, we have a second-order set of differential equations, and the effective driving force F_K acts directly only on the active charge component. This force and the coupling coefficients γ_{ij} entering the left-hand side of this system are given by (compare with Refs. 5 and 10)

$$F_K = m\tilde{\gamma}_p N_t \xi_e/2,$$

$$\gamma_{11} = \tilde{\gamma}_p(1+\xi_e), \quad \gamma_{12} = \tilde{\gamma}_e,$$

$$\gamma_{21} = \gamma_H, \quad \gamma_{22} = \gamma_H(1+\xi_h). \tag{6}$$

Here $N_t = N_C(N_\Sigma - N_C)/N_\Sigma$ is the effective trap concentration, $\gamma_H = e\mu_H H_0/\epsilon\epsilon_0$ is the rate of dielectric relaxation for the passive (compensating) carriers, $\epsilon\epsilon_0$ is the static dielectric constant, $\tilde{\gamma}_p = \gamma_p/(1+K^2L_D^2)$ is the renormalized rate of dielectric relaxation for photoexcited carriers with $\gamma_p = e\mu_p p_0/\epsilon\epsilon_0$ and $L_D = \sqrt{D_p\tau_p}$ the diffusion length, and $\xi_{p,H} = K^2R_{p,H}^2$ are dimensionless factors where $R_p = (\epsilon\epsilon_0 k_B T/N_t e^2)^{1/2}$ and $R_H = (\epsilon\epsilon_0 k_B T/H_0 e^2)^{1/2}$ are the Debye screening lengths.[13] Note also that $R_H/R_p = (N_t/H_0)^{1/2}$; the ratio N_t/H_0 and the product KR_p can thus be used as independent parameters instead of $KR_{p,H}$. The effective trap concentration N_t can generally depend on the sample history, e.g., on preillumination of the sample.[1–3]

It is worthwhile to mention that set (5), including its coefficients γ_{ij}, differs from the corresponding set (19) of Ref. 5 only by notation if the compensating carriers in the latter are set to be passive.

The field amplitude E_K is algebraically expressed in terms of the concentration amplitudes N_K and H_K:

$$E_K = ie(N_K + H_K)/\epsilon\epsilon_0 K. \tag{7}$$

Since the refractive index amplitude $\Delta n_K = -n^3 r E_K/2$ and the quantities n and r are known or measurable, expressing E_K in terms of the light amplitudes $A_{1,2}$ (or, more specifically, by the modulation coefficient m) is necessary to describe the observable optical effects. In particular, the diffraction efficiency of the index grating η is proportional to $|E_K|^2$ for samples not too thick, and the two-wave coupling gain factor Γ is proportional to $\mathrm{Im}(E_K/m)$ for samples not too thick or in steady state.[6]

4. FAST AND SLOW RELAXATION TIMES

Since the dynamical system has two degrees of freedom, it can be characterized by two relaxation times (rates). To determine these times, we set $F_K = 0$ and $N_K, H_K \propto \exp(-t/\tau)$. Then we have for τ^{-1} from Eqs. (5):

$$\tau_{f,s}^{-1} = \tfrac{1}{2}[\gamma_{11} + \gamma_{22} \pm \sqrt{(\gamma_{11}-\gamma_{22})^2 + 4\gamma_{12}\gamma_{21}}]. \tag{8}$$

The fast (f) and slow (s) relaxation times $\tau_{f,s}$ correspond to the signs (+) and (–), respectively.

The most important limiting case for SPS crystals is $\tilde{\gamma}_p \gg \gamma_H$; the factors $\xi_{p,H} = K^2R_{p,H}^2$ can generally be smaller than or of the order of 1. We obtain here from Eqs. (6) and (8):

$$\tau_f^{-1} \simeq \tilde{\gamma}_p(1+\xi_p), \quad \tau_s^{-1} \simeq \gamma_H \frac{\xi_p\xi_H + \xi_p + \xi_H}{1+\xi_p}. \tag{9}$$

Obviously, we have $\tau_s \gg \tau_f$. The value of τ_f is determined only by the characteristics of the active component; it coincides with the relaxation time calculated within the standard one-species mode[6] and complies with Ref. 5. The K dependence of τ_f is weak for sufficiently small and large values of the spatial frequency K. The slow time τ_s is de-

termined by the characteristics of both active and passive charge components; this is the fingerprint of strong charge-coupling effects. The difference between the rates τ_f^{-1} and τ_s^{-1} goes far beyond the difference between $\tilde{\gamma}_p$ and γ_H. In particular, the K dependence of τ_s is always strong for small values of the spatial frequency. Our result for τ_s differs essentially from that given by Eq. (25) of Ref.5.

One more limiting case is $\xi_{p,H} \ll 1$. We have here

$$\tau_f^{-1} \simeq \tilde{\gamma}_p + \gamma_H, \quad \tau_s^{-1} \simeq \frac{\tilde{\gamma}_p \gamma_H}{\tilde{\gamma}_p + \gamma_H}(\xi_p + \xi_H), \tag{10}$$

and again $\tau_s \gg \tau_f$. The fast rate τ_f^{-1} is the total rate of the dielectric relaxation, while the slow rate τ_s^{-1} with the strong K dependence is the classical rate of ambipolar diffusion.[8] The above-considered two limiting cases are overlapping for $\gamma_H \ll \gamma_p$, $\xi_{p,H} \ll 1$. The second case is especially important for the description of the fixing phenomenon in $LiNbO_3$ crystals.[10]

5. RECORDING KINETICS

As we know, the optical manifestations of the index grating are determined by the field amplitude E_K. However, the true state of the system and its evolution are determined by two concentration amplitudes N_K and H_K. Initial states with the same values of E_K but different combinations of N_K and H_K possess different subsequent field evolutions. It is necessary, therefore, to specify carefully the PR process under study.

We consider here the buildup of the grating with zero initial values of N_K and H_K for the frequency-degenerate case, $\Omega = 0$. It corresponds to the most typical conditions for experiments on PR recording. Since Eqs. (5) are linear, the buildup occurs with the rates τ_f^{-1} and τ_s^{-1}. In other words, any variable can be presented as a sum of fast and slow components; it does not mean that each of these components is attributed to a single (active or passive) type of charge carrier. For the space-charge field amplitude we have $E_K = E_f + E_s$, and the fast (f) and slow (s) field components obey the relations

$$E_{f,s} = E_{f,s}^0 (1 - e^{-t/\tau_{f,s}}), \tag{11}$$

where $E_{f,s}^0$ are the corresponding steady-state amplitudes. One can find then from Eqs. (5) and (6) under the condition $\gamma_H \ll \tilde{\gamma}_p$ that

$$E_f^0 \simeq \frac{imE_D}{2}\frac{1}{1+\xi_p},$$

$$E_s^0 \simeq -\frac{imE_D}{2}\frac{\xi_p}{(1+\xi_p)(\xi_p\xi_H + \xi_p\xi_H)}, \tag{12}$$

where $E_D = Kk_BT/e$ is the characteristic diffusion field. Both the amplitudes E_f and E_s are pure imaginary; i.e., the field grating remains $\pi/2$ shifted with respect to the light pattern during the whole recording process. The steady-state amplitude E_f^0 is not different from that known for the diffusion recording within the standard one-species model.[6] This means that the charge compensation is practically absent during the initial stage, $t \lesssim \tau_f \simeq \tilde{\gamma}_p^{-1}$. It is important that the slow steady-state amplitude E_s^0 is opposite in sign to E_f^0. This means that the total field amplitude $E_K = E_f + E_s$ decreases during the slow stage, after an initial fast increase. The ratio of the steady-state amplitudes is $E_f^0/E_s^0 = -(1 + \xi_H + N_t/H_0)$. The field compensation during the slow stage of recording is almost complete when $\xi_H = K^2R_H^2 \ll 1$ and $N_t/H_0 \ll 1$.

It is worth mentioning that the two-wave gain factors measured after completion of the fast stage and in the steady state are proportional to $\mathrm{Im}\, E_f^0/m$ and $\mathrm{Im}(E_f^0 + E_s^0)/m$, respectively. The corresponding values of the diffraction efficiency of the grating are determined by $|E_f^0|^2$ and $|E_f^0 + E_s^0|^2$, respectively.

It is also important to characterize the concentration amplitudes N_K and H_K during the recording process. The amplitude $N_K(t)$ grows monotonically during the f and s stages and reaches the steady-state value $N_K^0 = (m/2)N_t[1 + \xi_H/(\xi_p(1+\xi_H))]^{-1}$, which does not exceed $mN_t/2$. The amplitude $H_K(t)$, which is not driven directly by the force F_K, grows practically only during the s stage. For $t \gtrsim \tau_s$ it approaches the steady-state value $H_K^0 = -(m/2)N_t\xi_p/(\xi_p\xi_H + \xi_p + \xi_H)$; one can check that $|H_K| < mH_0/2$. The signs of N_K^0 and H_K^0 are opposite. Furthermore, the steady-state values of $|N_K|$ and $|H_K|$ are much larger than the values of these parameters achieved during the fast stage. Strong charge compensation therefore takes place during the slow stage of the grating recording.

6. FREQUENCY RESPONSE

Consider now the case $m \propto \exp(-i\Omega t)$, where Ω is the frequency detuning between the two recording light waves. It corresponds to the light interference pattern moving with velocity Ω/K. The steady-state value of the field amplitude is given here by

$$E_K^0 = \frac{imE_D}{2}\frac{\tilde{\gamma}_p(\gamma_H\xi_H - i\Omega)}{(\tau_f^{-1} - i\Omega)(\tau_s^{-1} - i\Omega)}. \tag{13}$$

At $\Omega = 0$, i.e., in the frequency-degenerate case, we obtain, using expressions (9),

$$E_K^0 = \frac{imE_D}{2}\frac{1}{1 + \xi_p + H_0/N_t}. \tag{14}$$

In the region $\tau_s^{-1} \ll \Omega \ll \tau_f^{-1}$ we have, instead,

$$E_K^0 = \frac{imE_D}{2}\frac{1}{1+\xi_p}. \tag{15}$$

For $\Omega\tau_f \gtrsim 1$, the function $E_K^0(\Omega)$ is monotononically decreasing.

The question is whether the value of E_K^0 given by Eq. (15) can be much larger than that given by Eq. (14). The only condition for that is the inequality $H_0/N_t \gg 1 + K^2R_p^2$. It can be easily fulfilled when $H_0 \gg N_t$. The frequency dependences of $|E_K(\Omega)|$ and $\mathrm{Im}\, E_K(\Omega)$ are characterized then by a narrow dip at $\Omega = 0$, followed by a plateau in the region $\tau_s^{-1} \lesssim |\Omega| \lesssim \tau_f^{-1}$ and a decreasing tail for $|\Omega|\tau_f > 1$. Note that the condition for the deep dip coincides with the condition of strong charge compensation during recording.

7. COMPARISON WITH EXPERIMENT AND DISCUSSION

Our main purposes here are (i) to demonstrate exemplarily that the theory is in a good qualitative agreement with experiment and (ii) to show how to determine the main material parameters of SPS from the standard set of experimental data. Quantitative determination of material parameters for particular samples is beyond this paper; this task would require a complete set of experimental data for each sample.

The recording kinetics of $|E_K|$ and $\mathrm{Im}\,E_K$ consists clearly of fast and slow stages, and the fast stage obeys the standard one-species model. Therefore the electronic (hole) parameters—the effective trap concentration N_t, the Debye and diffusion lengths R_p and L_D, and the dielectric relaxation rate γ_p—can be determined from the dependences of the maximum gain factor $\Gamma_f \propto \mathrm{Im}\,E_f^0$ and of the fast relaxation time τ_f on the grating spacing $\Lambda = 2\pi/K$, as was done, e.g., in Refs. 3, 12, and 14.

Figure 1 shows the dependence of the gain factor Γ_f^0 on the grating spacing; a similar dependence holds true for the square root of the diffraction efficiency $(\eta_f^0)^{1/2}$—both these quantities are proportional to $|E_f^0|$. The position of the maximum allows one to determine R_p according to expressions (12) for E_f^0. The values of R_p range approximately from 0.2 to 0.6 μm in SPS crystals.[13] It is necessary therefore to use both the transmission and the reflection geometries to proceed from small values of KR_p to values considerably larger than 1.

Figure 2 gives a representative K dependence of the fast relaxation rate (which is easily measurable in experiments) for the ratio of the diffusion and electronic Debye lengths $L_D/R_p = 2$. Behavior of $\tau_f^{-1}(K)$ within the first plateau section ($KR_p \lesssim 1$) allows one to estimate the value of the dielectric relaxation rate γ_p, and the ratio of the limiting values of τ_f^{-1} at low and high spatial frequencies gives the ratio L_D^2/R_p^2 in accordance with expressions (9) for τ_f. In experiment, the ratio L_D/R_p ranges from ≈ 1.5 to ≈ 10.[3,7,14]

Now we turn to the predictions that are beyond the standard model. One of them is the strong K dependence of the slow relaxation rate τ_s^{-1}. According to expressions (9), it can be represented in the following explicit form:

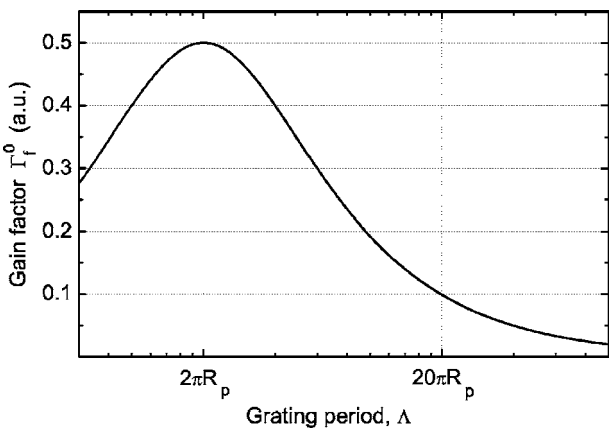

Fig. 1. Dependence of the maximum gain factor Γ_f^0 on the grating spacing Λ.

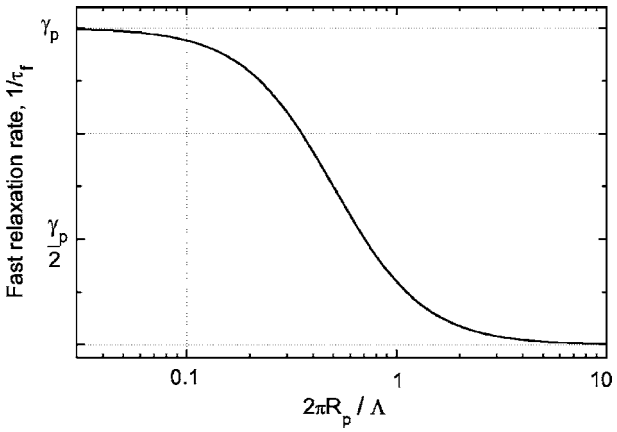

Fig. 2. Dependence of τ_f^{-1} on KR_p for $L_D/R_p = 2$.

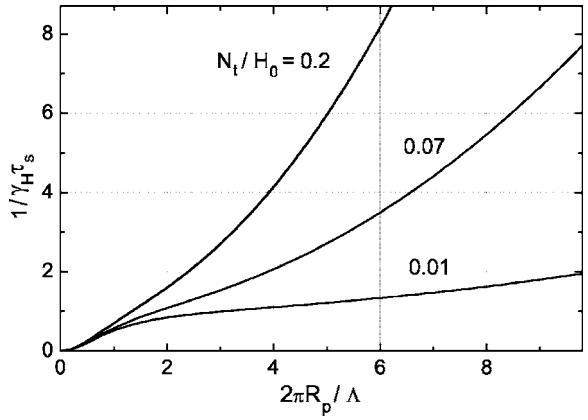

Fig. 3. Dependence of $1/\gamma_H\tau_s$ on KR_p for three representative values of N_t/H_0.

$$\tau_s^{-1} = \gamma_H\left(\frac{N_t}{H_0} + \frac{1}{1+K^2R_p^2}\right)K^2R_p^2. \tag{16}$$

For $KR_p \ll 1$, the decay rate τ_s^{-1} is proportional to $K^2R_p^2$, as one could expect for the ambipolar diffusion. This differs strongly from the prediction of Ref. 5 where $\tau_s^{-1}(K) = \mathrm{const}$.

Figure 3 shows the dependence of $(\tau_s\gamma_H)^{-1}$ on KR_p within the actual experimental range for $N_t/H_0 = 0.2$, 0.07, and 0.01. It is far from a quasi-constant one. The form of the K dependence essentially depends on the concentration ratio N_t/H_0. This form varies from a rapidly growing to a saturated one in the intermediate range of KR_p that is especially important for experiment. The initial section with the quadratic dependence is pretty narrow.

The increase of the slow decay time with the grating spacing observed experimentally in different samples[3,7] is in qualitative agreement with these predictions. However, the data measured at high spatial frequencies will be of primary importance to determine the values of H_0 and γ_H. To accomplish this task, one should include the data for reflection grating geometry in measured K dependence of the decay rate.

Consider now the recording kinetics. Its two key parameters are the ratios τ_f/τ_s and $(E_f^0+E_s^0)/E_f^0$. According to expressions (9) and (12), they can be represented as

$$\frac{E_f^0 + E_s^0}{E_f^0} = \frac{1 + K^2 R_p^2}{1 + K^2 R_p^2 + H_0 N_t^{-1}},$$

$$\frac{\tau_f}{\tau_s} = \frac{K^2 R_p^2 (1 + K^2 L_D^2)[1 + N_t H_0^{-1}(1 + K^2 R_p^2)]}{\gamma_p \gamma_H^{-1}(1 + K^2 R_p^2)^2}. \quad (17)$$

The condition of smallness of the first ratio (strong-field compensation in steady state) is obviously $H_0/N_t \gg 1 + K^2 R_p^2$. It is fulfilled for $N_t \ll H_0$ if $KR_p \lesssim 1$, but it can be violated for sufficiently large values of KR_p. The second ratio, τ_f/τ_s, is controlled by four dimensionless parameters: γ_H/γ_p, N_t/H_0, KR_p, and KL_D. For $KR_p \gtrsim 1$, it can be small only because of the smallness of γ_H/γ_p. Figure 4 shows a representative dependence of the refractive index amplitude (which is proportional to $|E_K| \equiv |\mathrm{Im}\, E_K|$) on the normalized recording time t/τ_f in a logarithmic scale. It corresponds to $\tau_f/\tau_s = 0.006$ and $E_f^0/(E_f^0 + E_s^0) = 6$. In the conventional linear time scale, the fast stage would not be easily distinguishable. One sees that the grating amplitude reaches the transient maximum at $t \simeq 4\tau_f$ and decreases then slowly to reach the steady-state value for $t \approx 500\tau_f \simeq 3\tau_s$.

The spectral dependence of the gain factor $\Gamma \propto \mathrm{Im}\, E_K(\Omega)$, given by Eq. (13), is presented in Fig. 5 for the same values of parameters. It is characterized by a narrow dip for $\Omega\tau_s \lesssim 1$, a plateau interval for $\Omega\tau_s \sim 10$, and a monotonically decreasing tail for $\Omega\tau_s \gtrsim 10^2$.

Qualitatively, all the above-shown dependences are in a good agreement with experiment. The gain spectra like that shown in Fig. 5 were first reported in Ref. 4, and their particular shape was calculated in Ref. 5. It should be underlined, however, that in the present paper the explicit expression for the phenomenological parameter τ_s is derived, see Eq. (16), and therefore the gain spectrum can be calculated. Alternatively, the found relations have a big potential for the determination and control of material parameters of SPS crystals (H_0, N_t, $\gamma_{p,H}$, $R_{P,H}$, and L_D) and also for shaping of the nonlinear response. The latter is important for applications of SPS crystals. For example, a strong K dependence of the characteristic slow decay rate can be used for adjusting the cutoff frequency in SPS-based novelty filters[15] and to manipulate the characteristics of the PR light slowing down.[16] In the last case,

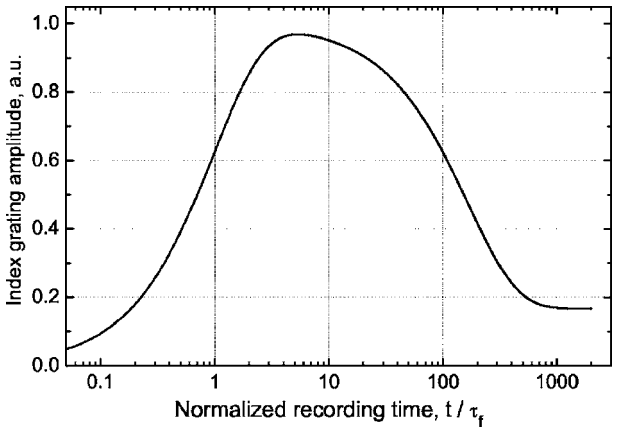

Fig. 4. Recording kinetics of $|\Delta n_K|$ for $KR_p = 1$, $L_D/R_p = 2$, $\gamma_p/\gamma_H = 250$, and $N_t/H_0 = 0.1$.

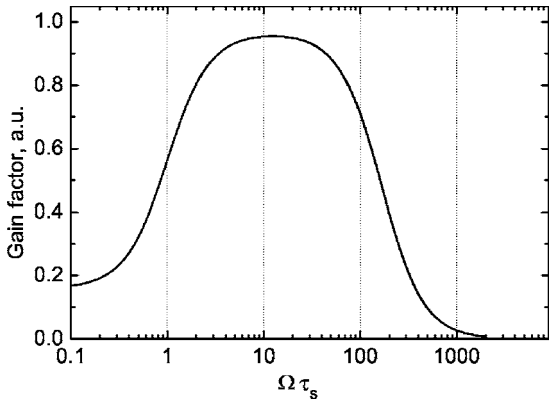

Fig. 5. Dependence of the gain factor Γ on the frequency detuning for the parameters of Fig. 4.

increasing the central dip in the spectral dependence of the PR response would improve the slow-light characteristics; equations of Section 6 predict how to control this dip.

It is interesting that our model automatically leads to a decreasing field amplitude $E_K(t)$ during the slow stage, which is consistent with experiments on two-wave coupling and grating diffraction.[1,3,17] One might think that this is the general feature of the charge compensation. However, it is not the case. As is known from the studies of the thermal fixing,[9,10] both scenarios (with increasing and decreasing E_K) are compatible generally with charge compensation.

An important advantage of the situation with the charge compensation phenomena in SPS over the situation with $LiNbO_3$ is relatively short values of the characteristic times $\tau_{f,s}$. In $LiNbO_3$ crystals, the slow time τ_s is often as long as weeks, which makes systematic studies of the K dependences in steady state complicated. This drawback is practically absent in SPS. This material can thus be considered one of the most suitable for the studies of charge compensation phenomena and shaping the relevant characteristics of the PR response.

Further development of the theory for SPS crystals can go in the direction of modeling of sophisticated erasure kinetics,[1,3] pulsating optical oscillation,[18] and novelty filters.[15]

8. CONCLUSIONS

We have developed a theory of the photorefractive response for $Sn_2P_2S_6$ crystals. It accounts self-consistently for the presence of active (photoexcited) and passive (compensating) charge carriers and takes into account specific material parameters of SPS. The theory explains well the main experimental observations, including two-step (fast–slow) recording kinetics, dependences of the fast and slow relaxation times on the grating spacing, and the spectral response with a narrow central dip. It has similarities with as well as important differences from the theory of thermal fixing in $LiNbO_3$ crystals. The theory rectifies the previous model considerations of the photorefractive response in SPS crystals and provides a tool for the characterization and shaping parameters of this important optical material.

The proposed theory can be applied to other photorefractive materials with two types of mobile charge carrier (optically active and passive) such as aluminium-doped bismuth titanium oxide,[19] and $Bi_4Ti_3O_{12}$.[20]

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