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Photorefraction of Pb-doped tin hypothiodiphosphate

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Abstract

Photorefractive properties of deliberately lead-doped tin hypothiodiphosphate ($Sn_2P_2S_6$, SPS) are investigated. In contrast to nominally undoped crystals, the space-charge gratings are formed by charge species of only one type, holes, what ensures a high beam-coupling gain in the steady-state. No optical sensitization is needed for the recording with near infrared light, but a photorefractive sensitivity does not extend beyond a wavelength of 0.9 μm. Like for some other SPS crystals, in SPS:Pb the effective trap density was found to be intensity dependent in the red spectral range.

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Keywords: Photorefraction of ferroelectrics; Charge transport and redistribution; Electrooptic effect

1. Introduction

Tin hypothiodiphospate ($Sn_2P_2S_6$, SPS) is known as a promising photorefractive material for the recording in the red and near infrared spectral range with high gain factor and rather short decay time (see, e.g., the review articles [1,2]). The largest amount of information is gathered so far for nominally undoped SPS crystals [2,3]. It has been shown that pre-exposure of the crystal to visible light with a quantum energy close to the forbidden band gap results in a considerable enhancement of the two-beam coupling gain factor at $\lambda = 1.06$ μm [3]. This was interpreted as a light-induced change of the effective trap density from $N_{eff} \approx 7.3 \times 10^{14}$ cm^{-3} for a virgin crystal to $N_{eff} \approx 7.7 \times 10^{15}$ cm^{-3} after pre-exposure [3]. Further, the effect of grating self enhancement was discovered when recording with He–Ne laser light with probably the same origin: Light with larger intensity ensured a larger gain factor [4]. The ultimate effective trap density measured in red light experiments was $N_{eff} \approx 10^{16}$ cm^{-3}.

These data allow to claim that the photorefractive efficiency of SPS can be considerably improved by technological means, e.g., via controlling the type and density of traps with appropriate impurities added during the growth procedure. Successful attempts in this direction have already been reported with Te and Sb-doped SPS crystals [5] and so called "modified" brown SPS crystals [6], especially in the near-infrared spectral range. The present paper describes the results of a photorefractive characterization of Pb-doped SPS crystals in the spectral range accessible with a Kr^+-laser and a Ti:Sapphire-laser. It is shown that this material ensures the steady-state two-beam coupling gain factor nearly as large as the steady-state gain factor determined for nominally undoped crystals, i.e., no compensating grating is revealed. A relatively high steady-state gain factor manifests itself in Pb-doped SPS with no preliminary exposure to white light, i.e., no sensitizing procedure is necessary as compared with nominally undoped crystals.

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2. Experimental procedure

The SPS crystals are grown by standard vapour-transport technique. Lead is loaded in the tube in the pure state with a concentration of about 1 wt.%. The SPS:Pb sample is cut along the crystallographic axes with dimensions $x \times y \times z = 5 \times 7 \times 3\ \mathrm{mm}^3$. Faces normal to the z-axis are optically finished. The results obtained are compared to that known for standard nominally undoped crystals ("yellow SPS" [1] or type-I SPS [4,7]).

The transmission-type dynamic gratings are recorded with cw-radiation of a TEM_{00} Kr^+-laser of wavelengths $\lambda = 0.568\ \mu$m (120 mW output power), $\lambda = 0.647\ \mu$m (400 mW), and TEM_{00} Ti:Sapphire-laser $\lambda = (0.75$–$0.9)\ \mu$m (700 mW). These two lasers, as it will be seen from what follows, cover nearly the whole range of photorefractive sensitivity of Pb-doped tin Hypothiodiphosphate. The unexpanded laser beam is used in all experiments. The light is polarized in the plane of beam intersection. The samples are aligned so that the photorefractive grating vector is parallel to the x-axis to profit from the presumably largest known (for undoped SPS) electrooptic coefficient $r_{111} = 174$ pm/V [8].

The dynamics of the two-beam coupling is studied in the transmission grating geometry. The measured quantities are the two-beam coupling gain [9]

$$\Gamma = \frac{1}{\ell} \ln \frac{I_s}{I_s(0)}, \tag{1}$$

with the interaction length ℓ and the intensity of the (weak) output signal wave I_s in the presence of the strong pump beam with the intensity $I_p \gg I_s$ and $I_s(0)$ without the pump wave and the characteristic exponential build-up (or decay) time of the refractive index grating τ. These quantities have been measured at various experimental parameters (grating spacing Λ, total light intensity I_0, intensity ratio I_s/I_p, light wavelength λ). From the experimental dependences we extract the Debye screening length that allows for evaluating the effective trap density, the diffusion length and therefore to estimate the lifetime-mobility product, the photo and dark conductivities, and the effective electrooptic constant that accounts for the degree of the poling. These data are compared to those known for nominally undoped material.

3. Experimental results and discussion

The temporal dynamics of the weak beam amplification which is due to beam coupling is shown in Fig. 1 for (*a*) SPS:Pb and (*b*) nominally undoped SPS of type I, respectively [4,7]. The dependences are quite different: As distinct for nominally undoped SPS [3], for lead-doped material the initially achieved high gain does not drop down with time. This points to a much smaller amplitude and much longer build-up time of the out-of-phase secondary grating [10] in SPS:Pb, if any grating of this kind exists at all. Qualitatively similar results have been obtained at all wavelengths from 0.568 μm till 0.9 μm. No beam coupling and no grating recording was observed in SPS:Pb at $\lambda = 1.06\ \mu$m. From the direction of the intensity transfer we conclude that within the whole range of photorefractive sensitivity the movable charge carriers in SPS:Pb are holes, similar to nominally undoped SPS [11].

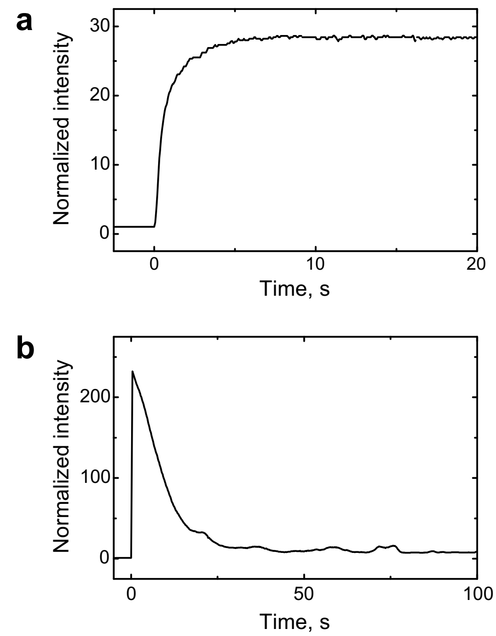

Fig. 1. Temporal evolution of the signal wave amplified because of two-beam coupling, (a) for SPS:Pb and (b) for nominally undoped type-I SPS. Pump-signal intensity ratio is 1000:1, total light intensity in the sample 1 W/cm^2, $\Lambda \approx 1.9\ \mu$m, $\lambda = 647$ nm.

The grating spacing dependences of the gain factor $\Gamma = \Gamma(\Lambda)$ are shown in Fig. 2 for different recording wavelengths. When measuring these dependences we kept a high intensity of the recording light >2 W/cm^2. The solid lines show best fits of the dependences calculated for crystals under the assumptions of one type of movable charge carriers [9]

$$\Gamma = \frac{4\pi^2 n^3 r_{\mathrm{eff}} k_B T}{\lambda \Lambda e \cos\theta} \cdot \frac{1}{1 + (\kappa I/\sigma_d)} \cdot \frac{1}{1 + K^2 \ell_s^2}, \tag{2}$$

to the experimental data. Here r_{eff} is the effective electrooptic coefficient, n is the refractive index, k_B is Boltzmann's

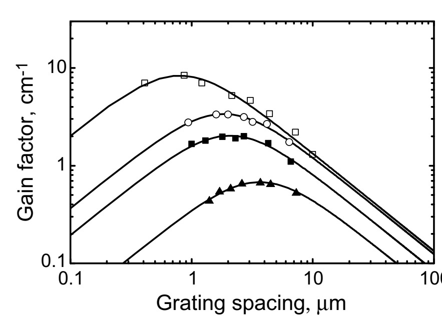

Fig. 2. Dependences of two-beam coupling gain factor Γ on grating spacing Λ for $\lambda = 0.568\ \mu$m (open squares), 0.800 μm (open dots), 0.850 μm (filled dots), and 0.875 μm (filled triangles).