Self-diffraction from two-photon absorption gratings in $Sn_2P_2S_6$

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Self-diffraction with the appearance of higher diffraction orders is discovered when writing a grating with a single sub-100 fs pulse in a nominally undoped $Sn_2P_2S_6$ sample. The short time of grating development, dependence of diffraction efficiency on the recording light intensity, correlation of wavelength dependence of efficiency with the spectrum of the two-photon absorption (TPA) constant, and a π phase shift of the diffracted beam allow for attributing the recorded grating to a dynamic amplitude grating of TPA.

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Tin hypothiodiphosphate ($Sn_2P_2S_6$, SPS) is a promising nonlinear material for dynamic holographic recording with cw light [1] as also with repetition rate pulsed radiation up to the picosecond range [2]. It has been shown recently that femtosecond light pulses with photon energies within the range from 1.5 to 2.1 eV are strongly attenuated in nominally undoped SPS crystals via two-photon absorption (TPA), with a TPA coefficient β that reaches a maximum value up to 8 cm/GW [3]. Thus, for the peak pulse intensities $I \approx 1\ \mathrm{GW/cm^2}$ the product $\beta I = 8\ \mathrm{cm^{-1}}$ is roughly 1 order of magnitude larger than the linear absorption of SPS $\alpha \leq 1\ \mathrm{cm^{-1}}$ [1] and the recording of high contrast amplitude gratings could be expected.

Apart from instantaneous amplitude gratings of nonlinear absorption the interfering femtosecond pulses may also record phase gratings in wide bandgap photorefractive crystals as SPS: an instantaneous index grating via optical Kerr effect and inertial index grating via excitation of free carriers or polarons [4,5] as also space charge grating [5,6]. Gratings related to other nonlinear effects can be expected, too. In this Letter, we report on dynamic grating recording with single femtosecond pulses and describe the studies that allow for identifying the process that contributes the most to the grating appearance.

The SPS crystals are grown in the Institute of Solid State Physics and Chemistry of Uzhgorod State University by chemical vapor transport. The most part of the measurements presented below are performed with an z-cut nominally undoped sample (sample identifier K16, with thickness $\ell = 6.5$ mm along z axis). A 1 kHz repetition rate tunable laser with sub-100 fs pulses (see Ref. [3]) is used as a light source (600–760 nm, $\bar{P} \leq 20\ \mu\mathrm{J}$). The pulse energy at the sample input face is controlled with a set of calibrated neutral density filters. The attenuation with relatively small temporal pulse broadening (roughly within 5%) is ensured.

To record a grating, a 20 lines/mm Ronchi ruling is imaged into the SPS sample with a symmetric telescope formed by two lenses L_1 and L_2 with the focal length $F_1 = 150$ mm (Fig. 1). The amplitude mask AM_1 is placed in the Fourier plane of the first lens L_1 that selects only $+1$ and -1 diffraction orders from the multitude of the diffraction spots. In such a way a high-contrast fringe pattern with cosine intensity distribution is projected inside the sample, with the fringe spacing $\Lambda = 25\ \mu\mathrm{m}$. This technique has been successfully used in the past for photorefractive grating recording with spatially incoherent light [7]. It appears to be especially attractive when writing gratings with femtosecond pulses [8] because of the best possible temporal and spatial overlap of short pulses.

Apparently, the self-diffraction from a homogeneous in depth dynamic grating with 25 μm spacing in a 6.5 mm thick sample should be well beyond Raman-Nath type as the relevant Klein-Cook factor $Q = (2\pi\lambda\ell/\Lambda^2 n) \approx 14$ is larger than 10 [9] and Moharam–Gaylord criterion [10], $\rho = (\lambda^2/n\Delta n\Lambda^2) \approx 22 \gg 10$ is also met with justified assumption that $\Delta n \leq 10^{-5}$. Several different phenomena can lead, however, to the appearance of the higher diffraction orders. The most plausible is in strong reduction of the effective depth ℓ_{eff} of the dynamic grating (up to $\approx 100\ \mu\mathrm{m}$ at $I \approx 10\ \mathrm{GW/cm^2}$) and consequent violation of the Klein-Cook criterion. Whatever the real reason is, higher diffraction orders, apart from the transmitted two recording beams, are clearly observed in the Fourier plane of the lens L_3 placed behind the SPS sample.

By selecting the first non-Bragg order [11] with the mask AM_2 the energy of the diffracted pulse can be measured with a Si-PIN detector (not shown in Fig. 1). Being normalized to the recording pulse energy it gives the measure of the integral diffraction efficiency η.

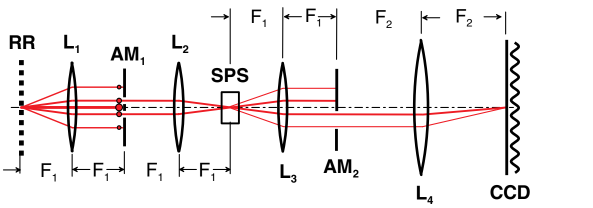

Fig. 1. (Color online) Sketch of the experimental setup with the telescope lenses L_i, amplitude masks AM_i, sample SPS, CCD line array, and Ronchi ruling RR.

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The lens L_3 is also a part of the second telescope, which is imaging the output surface of the SPS sample to the CCD line array (with magnification, as the focal length of the lens L_4 is $F_2 = 2250$ mm.) The intensity distribution along the wave vector of the fringe pattern is measured with the CCD line array and stored in a data acquisition system. The spatial filtering in the Fourier plane of the lens L_3 allows for passing the fringe patterns formed either by the two transmitted recording beams or by one recording and one diffracted beam to the CCD line array. By determining the phase shift between these two fringe patterns one can distinguish, as it is explained below, the type of the recorded grating (amplitude or phase grating).

In the first set of experiments we measure the diffraction efficiency η in individual pulses for an established repetition rate regime with 1 Hz frequency. No cumulative effects are detected within the power and energy range of pulses used for recording. After several minutes of continuous periodic excitation the amplitude of the diffracted signal remains the same as in the few initial pulses, fluctuating within ±5%. This allows for averaging the measured data over the train of consecutive 20 pulses, to reduce their natural scatter.

Then, the intensity dependence of the diffraction efficiency for different recording wavelengths is measured (Fig. 2). The intensity was calculated under approximation of a Gaussian temporal profile; all data refer to the peak intensity values. Within the error bars all dependences of Fig. 2 show roughly a linear increase of the efficiency versus intensity, $\eta \propto I$.

Taking into account that η is an average in time and in space but not local and instantaneous value of the diffraction efficiency the linear $\eta \propto I$ dependence in Fig. 2 has no special meaning here and is most probably accidental, typical only for limited intensity range in which our measurements are done. It allows nevertheless to claim that we are dealing with the nonlinear recording and extract from the data of Fig. 2 the normalized spectral sensitivity of SPS to grating recording.

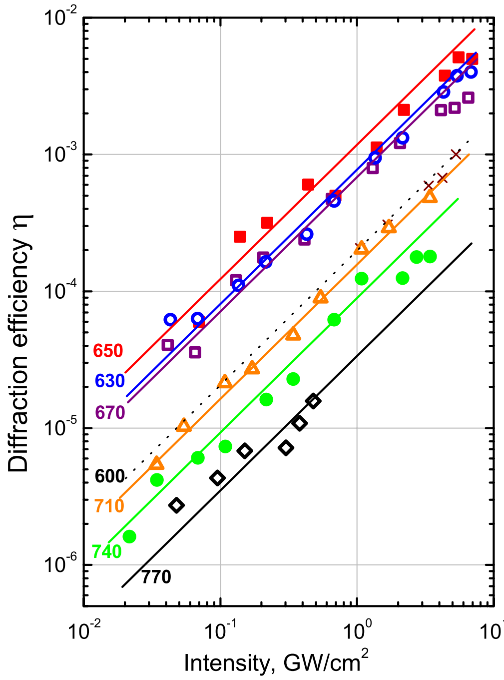

Fig. 2. (Color online) Intensity dependence of the diffraction efficiency for the recording wavelengths 600 nm (crosses), 630 nm (open dots), 650 nm (filled squares), 670 nm (open squares), 710 nm (triangles), 740 nm (filled dots), and 770 nm (diamonds). Solid lines are drawn to guide the eye.

Figure 3 shows the wavelength dependence of the diffraction efficiency η measured at $I = 1$ GW/cm^2. This dependence fits qualitatively well to the spectrum of the TPA coefficient β for nominally undoped SPS (see Fig. 3 of Ref. [3]), with its maximum in the vicinity of $\lambda = 650$ nm. It might suggest that our dynamic grating is somewhat related to nonlinear absorption in the sample and rule out its attribution to the optical Kerr effect. It does not prove, however, that self-diffraction originates just from the absorption grating itself: being excited via TPA the gratings of free carriers or polarons can demonstrate similar spectra, too.

While the TPA grating is a pure amplitude grating the free carrier and polaron gratings may feature both amplitude and phase components, with relative weights that depend on the excitation wavelength (see, e.g., the data of Ref. [12]). As it was shown in [3] the contribution of free carriers or polarons to the nonlinear absorption in SPS is detectable but very small, less than 7% of TPA at 630 nm. At the same time, even relatively small index variation can result in noticeable diffraction. Thus, for identifying the dominant process responsible for self-diffraction one needs to establish the type of the appearing grating: amplitude, phase, or mixed grating.

It is known [10] that for the phase grating the phase difference between the adjacent orders is $\pi/2$, while for the amplitude grating this difference is either zero or π. With this in mind, we compare the relative positions of two fringe patterns, one generated by two transmitted recording beams and the other from one recording beam and the adjacent diffracted beam (as it is shown in Fig. 1). The selection of relevant diffraction orders is made in the focal plane of the lens L_3 with aperture AM_2. The recorded intensity distributions are shown in Fig. 4(a) by dashed black and solid color lines. The sample optical imperfections affect the fringe quality, especially for

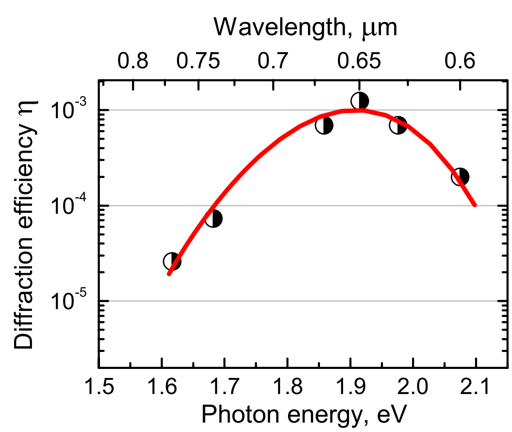

Fig. 3. (Color online) Diffraction efficiency of the dynamic grating in the 6.5 mm thick SPS sample versus recording wavelength for a fixed recording intensity of 1 GW/cm^2. Solid line is drawn to guide the eye.

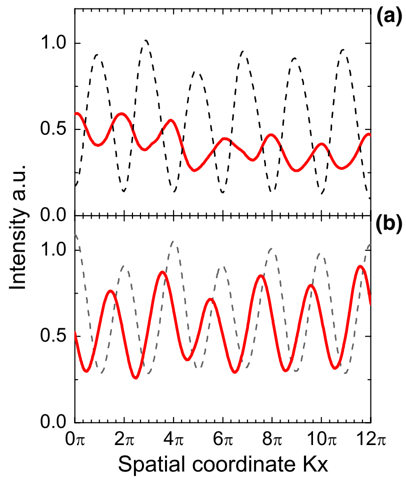

Fig. 4. (Color online) Intensity scan of the fringe pattern from the two recording light beams (dashed line) and from the recording and non-Bragg diffracted beams (solid line). (a) Grating recording and simultaneous readout with 75 ps light pulses at $\lambda = 650$ nm in the 6.5 mm thick SPS sample. (b) Grating recording and readout with cw He–Ne laser light ($\lambda = 630$ nm) in a 0.5 mm thick SPS sample.

low contrast fringes produced by the recording and diffracted beams. This does not prevent, however, from concluding that in both cases the fringes with the same spacing are observed and that *they are nearly out-of-phase, i.e., fringes are π-shifted.*

To check the validity of such a technique of phase shift evaluation a similar test is done with cw He–Ne laser radiation. It is expected that the dominant dynamic grating in case of cw recording is the phase grating, which is due to space charge redistribution [1].

The whole recording arrangement is kept the same except the laser source and SPS sample. The observation of non-Bragg diffraction orders appeared to be impossible with the 6.5 mm thick sample. Therefore, a thin z-cut sample of nominally undoped SPS with $\ell = 0.5$ mm is used, its x axis being aligned perpendicularly to the light fringes. The Klein-Cook factor is small enough, $Q \approx 1.3$, to observe Raman-Nath self-diffraction from the thin photorefractive grating.

Figure 4(b) shows the results of the measurements, with the same color coding, dashed black line for recording fringes and solid color line for fringes from the recording beam and the adjacent non-Bragg diffracted beams. The difference to the result presented in Fig. 4(a) is evident: now two fringe patterns are dephased roughly to one-quarter of fringe spacing, thus indicating a $\pi/2$-shift between the adjacent Bragg and first non-Bragg orders.

All results mentioned above lead to the conclusion that the most significant part of the described self-diffraction of femtosecond pulse radiation in SPS comes from the dynamic amplitude grating related to two photon absorption.

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11. We use these notations to distinguish between "Bragg" orders (two recording beams being Bragg-matched to the fundamental grating with the grating vector $K = k_1 - k_2$) and two adjacent, "non-Bragg", orders (i.e., violating Bragg condition for fundamental grating K). Note, that the recording beams are not Bragg-matched for the grating with the doubled spatial frequency $2K$, which also occurs under two-photon excitation.
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