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Spectral dependence of cross-talk between photorefractive gratings in $Bi_{12}SiO_{20}$ in diffusion regime

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Abstract. We report on experimental results showing a strong wavelength-dependence of cross-talk between photorefractive gratings simultaneously recorded in $Bi_{12}SiO_{20}$ crystals in the diffusion regime. We find unusually high cross-talk for two gratings with close spatial frequencies at wavelengths 488 and 476 nm. The results indicate that the density of free charge carriers does not directly follow light modulation at low spatial frequencies.

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In photorefractive crystals, inhomogeneous illumination yields a charge redistribution, a space-charge field builds up and modulates the refractive index via the electrooptic effect [1]. This effect may be used in many applications. For several devices superposition of many angular-multiplexed holograms within the same volume is required, e.g. in volume holographic storage [2]. Therefore cross-talk, or the influence of recording and erasure of one grating on the diffraction efficiency of another grating, is a substantial problem which limits the performance of photorefractive devices. Recent studies of nonlinear combinations of photorefractive gratings have demonstrated a strong coupling between coherent gratings [3–6].

In this paper we report that new measurements with light of shorter wavelength (488 and 476 nm from an Ar^+ laser) reveal even greater cross-talk than our previous measurements at the green line 514.5 nm [4–6].

Our measurements were performed with photorefractive bismuth silicate $Bi_{12}SiO_{20}$ (BSO) because this material has fast response times for the usual intensities of cw laser light and high enough diffraction efficiency at the diffusion regime.

1 Theoretical background

According to the well-known PDDT model (photogeneration, diffusion, drift, trapping), or band transport model [7] the photorefractive effect responsible for the recording of phase gratings does not directly depend on the wavelength of the writing waves. The photon absorption cross section, which is multiplied with the total light intensity to describe the generation rate of free carriers, may have an influence only on time constants of the space-charge build-up. However, we find experimentally that the cross-talk between neighboring simultaneously recorded gratings in BSO depends strongly on the wavelength of the writing beams.

We study cross-talk in a three-wave mixing configuration with two object beams, O_1 and O_2, of equal intensities, I_1 and I_2, and small angular separation and a reference beam R with intensity I_R. In a BSO crystal without applied voltage (diffusion regime) the beams O_1 and R record the grating G_1, and O_2 and R record the grating G_2. The corresponding grating spacings are Λ_{1R} and Λ_{2R}. The two object beams O_1 and O_2 are unable to create a sizeable grating in the diffusion regime due to the very large spacing Λ_{12} of the interference between them. The angle between the object beams and the reference beam is adjusted for efficient holographic grating formation.

The origin of cross-talk between the gratings may be explained as follows: each primary grating is recorded by the reference beam and the corresponding object beam. Even though the light intensity pattern produced by the interference between the object beams does not produce any detectable grating, it modulates the local generation of free electrons with a low spatial frequency. This periodic spatial modulation causes the formation of a space-charge field with new spatial frequencies separated by the difference wave number $\Delta K = K_{1R} - K_{2R}$, where $K_{1R} = 2\pi/\Lambda_{1R}$ and $K_{2R} = 2\pi/\Lambda_{2R}$ are the wave numbers of the gratings G_1 and G_2 (Fig. 1) [5]. The spectral power is now spread out over a wider range of frequencies. The periodic space-charge field with spatial frequency K_{1R} is, therefore, a combination of the central spectral component associated with grating G_1 and a side lobe of grating G_2 spectrum. This coupling through the low spatial frequency electron distribution is the reason for the cross-talk between the gratings, which can be measured experimentally as a change in diffraction efficiency. We define the cross-talk $\Delta\eta$ as $\Delta\eta = (\eta_a - \eta_b)/\eta_b$, where η_b and η_a represent the

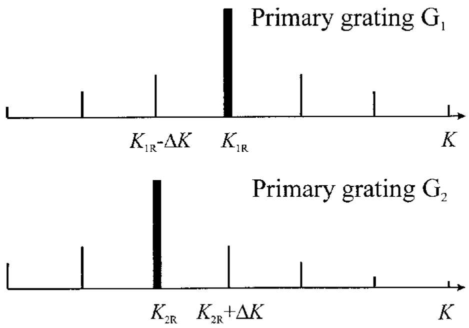

Fig. 1. Spatial spectra of two neighboring gratings G_1 and G_2 showing the appearance of additional frequencies caused by light modulation with spatial frequency ΔK

steady-state diffraction efficiencies of the grating G_1 *before* and *after* grating G_2 is 'switched off' by means of an appropriate phase-modulation of beam O_2 [3].

With the often-used assumption that the electron density has the same form as the incident light intensity, apart from a constant reduction factor, we can express the space-charge field along the x axis, $E_{SC}(x)$, when both gratings exist in the crystal, as follows:

$$E_{SC}(x) = \frac{k_B T}{e} \cdot \frac{M_{1R} K_{1R} \sin(K_{1R} x) + M_{2R} K_{2R} \sin(K_{2R} x)}{1 + M_{12}\cos(\Delta K x) + M_{1R}\cos(K_{1R} x) + M_{2R}\cos(K_{2R} x)}, \quad (1)$$

and when only one grating G_1 exists in a crystal, (1) becomes

$$E_{SC}(x) = \frac{k_B T}{e} \frac{M_{1R} K_{1R} \sin(K_{1R} x)}{1 + M_{1R}\cos(K_{1R} x)}, \quad (2)$$

where M_{ij} are modulation depths of the free electron density: $M_{ij} = m_{ij}(1-\delta_{ij})/(1+b_t/sI_0)$, $m_{ij} = 2\sqrt{I_i J_j}/I_0$ are the light-intensity modulation depths, I_1, I_2 and I_R are the intensities of object and reference beams respectively, $I_0 = I_1 + I_2 + I_R$ is the total intensity of the incident light, δ_{ij} is the reduction factor for the carrier modulation, k_B is Boltzmann's constant, T is the absolute temperature, e is the electron charge, sI_0 is the rate of free-carrier photogeneration and b_t is the rate of thermal excitation of carriers. The expression $(1-\delta_{1R}) = 1/(1+K_{1R}^2/K_D^2)$ for the reduction is used to describe the limitation in diffraction due to the maximum achievable space-charge field [7]. Here K_D denotes the Debye screening wave number, $K_D^2 = e^2 N_A/\varepsilon\varepsilon_0 k_B T$, where N_A is the concentration of acceptors and $\varepsilon\varepsilon_0$ is the permittivity of the crystal. The value of K_D can be determined from the dependence of diffraction efficiency on the spatial frequency K_{1R} of the grating.

There will be hardly any reduction of the modulation factor for low-frequency carrier distribution due to the large period of grating G_{12}. However, without a reduction factor the expression (1) may contain a singularity. In our previous work we used a fitting procedure to select the value of the reduction factor $1-\delta_{12}$, which gives the best fit to experimental data [5].

To calculate the cross-talk $\Delta\eta$ numerically, we use the Fourier transform of the relations (1) and (2) to compute the fundamental spatial frequency component of E_{SC} at the wave number K_{1R}. Thus, we obtain values of the strength of grating G_1 before and after switching off grating G_2. The squares of these values are proportional to diffraction efficiencies η_b and η_a, and hence the cross-talk $\Delta\eta$ may be calculated.

The presented model of grating coupling via low-frequency modulation neglects the possibility of energy transfer between the object beams by diffractive coupling through the reference beam. The beam coupling in BSO in the diffusion regime is usually small, and its influence was found to be of the order of 1% [5].

2 Experimental part

The experimental recording configuration is shown in Fig. 2. Three collimated plane waves O_1, O_2 and R are incident on the BSO crystal and fully cover its entrance face. The object beams are separated by a small angle $\Delta\theta$ of between 1 to 10 mrad. The angle between the reference beam and the object beams θ is about 30°. All three beams originate from a single mode Ar^+-ion laser with polarization perpendicular to the plane of incidence. In the experiments we use the laser lines at 514.5, 488 and 476 nm. A cw frequency-doubled Nd:YAG laser at wavelength 532 nm is also used in the same scheme. The intensities of all beams are measured in front of the BSO crystal by a power meter. A He-Ne laser beam at wavelength 633 nm is used to measure grating diffraction efficiencies. The angle of incidence of the He-Ne laser probe beam is Bragg-matched to the spatial period Λ_{1R} of grating G_1, which is about 1 μm.

A phase-modulation was imparted on one of the object beams by means of a mirror mounted on a piezoelectric stack. By a special choice of frequency and amplitude the phase-modulation will completely erase the grating G_2 without changing the total intensity of light incident on the crystal [3, 5]. The typical value for the frequency of phase-modulation is 300 Hz and the modulation amplitude corresponds to the first zero of the Bessel function [5]. The cross-talk was measured for different values of the parameter $\beta = I_R/(I_1 + I_2)$, which

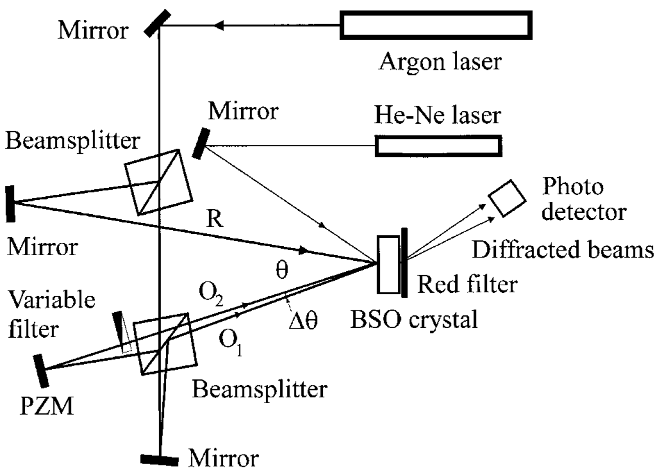

Fig. 2. Experimental set-up for the cross-talk measurements. Two object beams O_1 and O_2 with equal intensities and small angular separation $\Delta\theta = 3$ mrad write gratings with a reference beam R. Beam O_2 can be phase-modulated by a piezoelectrically supported mirror (PZM). The beam intensity ratio $\beta = I_R/(I_1 + I_2)$ is controlled by a variable neutral density filter

describes the intensity ratio between reference beam and the object beams.

Three different BSO crystals of thicknesses 3, 5 and 10 mm are investigated. The surfaces are polished to optical quality and antireflection-coated to avoid build-up of additional gratings by interference from internally reflected light beams. The measured value of K_D in the two-beam configuration reveals $K_D \approx 10\,\mu m^{-1}$. With the $K_{1R} = 6.3\,\mu m^{-1}$ used in three-beam experiments a reduction factor $1 - \delta_{1R} = 0.72$ follows.

Figure 3 represents the dependence of the cross-talk on the intensity ratio β obtained for four different wavelengths. The difference between the curves is most evident in the region of small β, where gratings G_1 and G_2 have a low modulation index ($m_{1R} = m_{2R} \ll 1$) and the influence of the low spatial frequency electron distribution is at a maximum (m_{12} is close to unity). The measured cross-talk for low values of β depends greatly on the wavelength, and at 476 nm is 2–3 times higher than at 514.5 nm.

We have not observed any diffraction indicating the existence of grating G_{12} at any available wavelength. The grating with wave vector $\Delta K = K_1 - K_2$ does not exist at all in the diffusion regime, but it is easily detectable with applied external voltage.

Additional measurements were performed to ascertain that the difference is only due to the difference in wavelength. The cross-talk was measured for different values of the total intensity, but no significant variation of the cross-talk was observed. As a further check, the cross-talk was measured at different wavelengths for the same value of the photogeneration rate, measured by the photocurrent with uniform illumination and an external applied electric field of 1 kV/cm. The results for the cross-talk were the same as shown in Fig. 3.

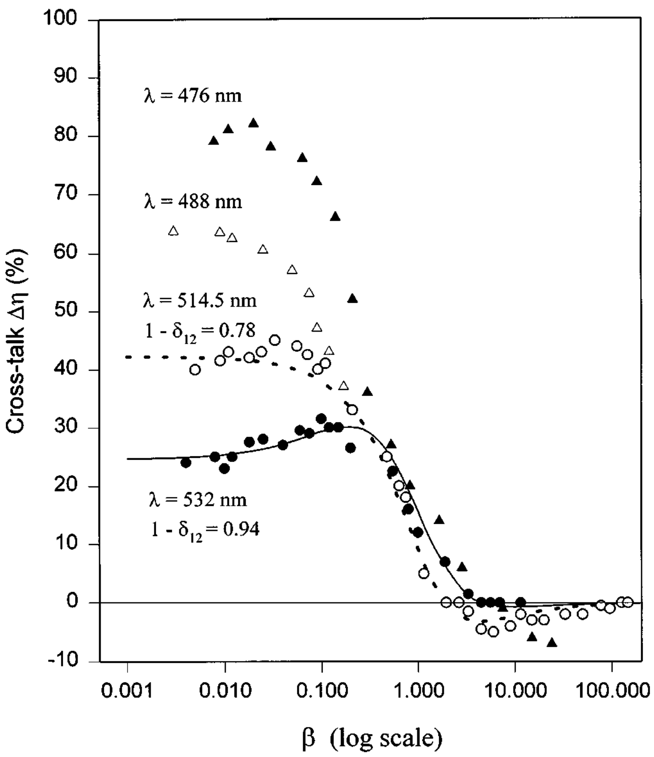

Fig. 3. Measured cross-talk $\Delta\eta$ for four wavelengths as a function of the intensity ratio β. *Filled circles*: $\lambda = 532$ nm. *Solid curve*: calculation with a fitting parameter $\delta_{12} = 0.06$. *Open circles*: $\lambda = 514.5$ nm. *Dashed curve*: calculation with a fitting parameter $\delta_{12} = 0.22$. *Open triangles*: $\lambda = 488$ nm. *Closed triangles*: $\lambda = 476$ nm

Measurements at a β value of about 0.01 give the same cross-talk for all three tested crystals indicating that the cross-talk is independent of changes in the optical path length through the crystal and of small differences in the material parameters of the crystals.

Furthermore, we tested the influence of changes of the angular separation $\Delta\theta$ between the object beams ranging from 1 to 10 mrad. The dependence on $\Delta\theta$ is small, in agreement with earlier results [5]. Thus, the relatively small changes of the grating period, resulting from changes of the wavelengths, are not the origin of the difference in the measured cross-talk.

Finally, the modulation depth of the interference pattern at the different wavelengths was checked. We measured the light modulation index to be 0.96 at 514.5 nm and 0.92 at 476 nm. This was done by using a photomultiplier with a narrow slit-aperture at a magnification of 25 times interference pattern image. This difference is much too small to explain the difference in cross-talk at these two wavelengths.

3 Discussion

Our measurements at 514.5 and 532 nm agree well with the theoretical expression for the space-charge field for the three incident beams presented in [4, 5], when an appropriate value for the reduction factor $1 - \delta_{12}$ is chosen, as shown by the theoretical curves in Fig. 3. However, the high values of cross-talk measured at the 476 and 488 nm Ar^+-laser lines cannot be fit with any choice of the reduction factor. This indicates that additional physical effects must be included in the analysis to account for the experimental observations at 488 and 476 nm.

To explain the revealed cross-talk for $\lambda < 500$ nm we assume that the density of free-charge carriers does not directly follow light modulation at low spatial frequencies. To perform the fitting procedure we approximate the corresponding form of the carrier distribution as: $1 + m(1 - \delta_{12})[\cos(\Delta Kx) + a\cos(2\Delta Kx) + b\cos(3\Delta Kx)]$. The value of calculated cross-talk is found to be very sensitive to this introduced nonlinearity of the free carrier distribution. The best fit for measured cross-talk at $\lambda = 476$ nm is obtained with $\delta_{12} = 0.2$, $a = -0.196$, and $b = 0.03$, as shown in Fig. 4. This result indicates that the nonlinearity grows for shorter wavelengths.

We have analyzed a number of physical factors to explain the spectral dependence of the cross-talk. As an example, short-wavelength light can generate non-thermalized electrons with surplus kinetic energy. Such non-thermalized electrons increase the effective diffusion constant, which can enlarge the nonlinearity at low spatial frequencies. Another possibility is activation of deep donor levels with the short-wavelength light [8]. A change in the excitation rate of donor levels or a filling of traps with the change of wavelength may lead to differences in the grating formation and in the nonlinear coupling between gratings. There are many indications that the charge transport in sillenites is governed by more than one photorefractive level [9]. Different centers or one center which occurs in more than two different valence states are possible origins of multiple photorefractive levels [10]. The

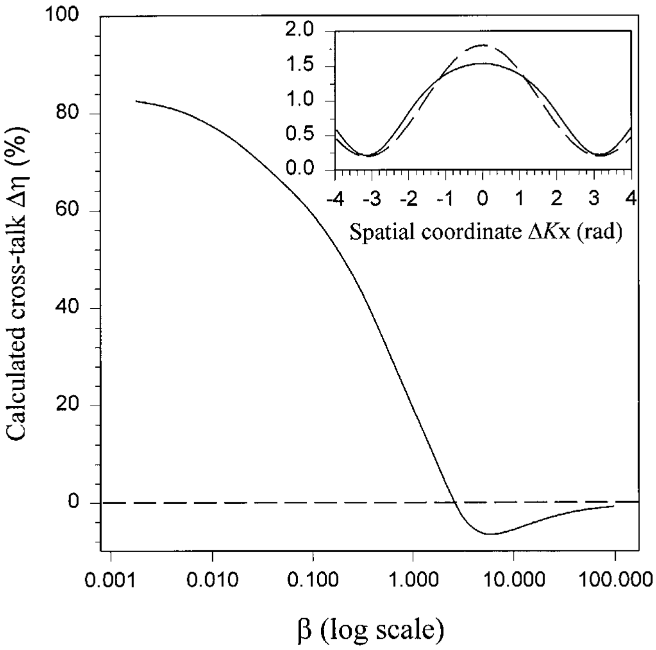

Fig. 4. Calculated cross-talk as a function of the intensity ratio β for a low-frequency distribution of free carriers of the form $1+m_{12}(1-\delta_{12})[cos(\Delta Kx)+a\cos(2\Delta Kx)+b\cos(3\Delta Kx)]$. *Insert*: distribution of free-charge carriers (*solid line*) in the form given already and modulation of light intensity (*dashed line*) in the form $1+m_{12}\cos(\Delta Kx)$, for $m_{12}=0.8$, $\delta_{12}=0.2$, $a=-0.196$ and $b=0.03$

degree of nonlinearity can depend on the light wavelength, according to the spectral dependence of the photoionization of cross sections of the different levels. Other possible explanations include the increased formation of absorption gratings at short wavelength [11], or a change in the balance between the generation of electrons and holes [12]. In general, if the photoconductivity increases sublinearly with light intensity a reduction of free-charge distribution appears [13]. One of possible mechanisms for cross-talk can also be the coupling between the gratings by means of high diffraction orders [14, 15]. We note, however, that the nonlinear response at high spatial frequency resulting in appearance of high diffraction orders produces very little influence on the cross-talk between gratings due to low spatial frequency modulation of carriers distribution as in our case. Our present data do not allow us to select a particular reason for the increased cross-talk in the short-wavelength region.

From our investigation it appears that in BSO crystals multiplexing of holograms with light of wavelengths shorter than 500 nm is disadvantageous because of enlarged cross-talk. Our study may be important for optical storage in photorefractive materials. The effect of cross-talk between close holographic gratings shows the limitations for optical storage in photorefractive media.

Here we have experimentally demonstrated the strong relationship of the cross-talk between two gratings on the wavelength in three-wave mixing experiments in BSO crystals in the diffusion regime. A strong cross-talk at 476 nm of about 100% has been measured. This experimental method of cross-talk measurement could become a very sensitive tool for the study of charge transport in photorefractive materials.

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