

RECORDING OF HOLOGRAPHIC GRATINGS AND THEIR COHERENT SELF-ENHANCEMENT IN AN a-As₂S₃ FILM WITH A MINIMUM LIGHT INTENSITY MODULATION

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The holographic grating recording efficiency and the coherent self-enhancement efficiency of gratings are experimentally studied depending on the recording light interference fringe visibility in an a-As₂S₃ chalcogenide film in order to find the minimum visibility. The minimum fringe visibility $M_{\min} = 3.6 \times 10^{-4}$ is found, which is determined by the scattered light background. In this case the maximal diffraction efficiency $\eta_{\max} = 0.05\%$ and the maximal self-enhancement factor $\xi_{\max} = 5.0$, compared to $\eta_{\max} = 15\%$ and $\xi_{\max} = 12.3$ in the optimal $M = 1.0000$ case. In the case of two-beam holographic grating recording the maximal diffraction efficiency increases when M is increased, whereas sensitivity decreases. A simple model, based on linear recording with the spatial light intensity distribution governed by M and including intensity-dependent material photosensitivity and grating relaxation, is proposed to explain these results. In the case of coherent self-enhancement both the maximal diffraction efficiency and sensitivity increase with fringe visibility M . This can be explained by the fact that mechanical stress modulation during the initial grating recording depending on M is followed by relaxational structural changes reinforcing the coherent self-enhancement effect. The developed approach may explain the absence of this effect in some azobenzene oligomer films.

Keywords: amorphous chalcogenide films, holographic gratings, hologram self-enhancement, interference fringe visibility

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1. Introduction

The efficiency of holographic information recording depends to a large extent on the recording light interference pattern fringe visibility M and on the modulation transfer function (MTF) of the recording material [1]. In the simplest case the information to be recorded contains only one spatial frequency. Then we have an elementary hologram – a holographic grating (HG). In HG spectroscopy HG serves as a tool for the studies of material properties. Usually HG spatial frequency is varied. Detailed studies of HG recording efficiency on fringe visibility are much less widespread. Yet such studies at a fixed spatial frequency can be regarded as a special case of holographic grating spectroscopy of materials. The aim of this work was to study the recording light interference

fringe visibility dependence of HG recording efficiency in an a-As₂S₃ film by two beams (as usual) and by one beam in the process of HG coherent self-enhancement [2, 3] focusing on extremely low visibilities.

HG recording intensity distribution along the x -axis can be described as

$$I = I_0 (1 + M \cos Kx), \quad (1)$$

where

$$M = 2(P_1 \cdot P_2)^{0.5} / (P_1 + P_2) \quad (2)$$

is fringe visibility, P_1 and P_2 are the recording beam powers, I_0 is the average light intensity, $K = 2\pi/\Lambda$ is HG angular spatial frequency, Λ is HG period. If $M = 0$, there is no recording.

Amorphous As₂S₃ semiconductor chalcogenide films are used for real-time holographic information storage, optical computing, and for making holographic optical elements [2-4]. Diffraction efficiency exceeding 80% [4], high spatial resolution of spatial frequencies up to 9700 lines/mm [4, 5], simplicity of a direct recording process for practically permanent holograms which need neither fixing nor development, reversibility, and availability of large samples are some of the attractive features of these materials. Holographic recording in a-As₂S₃ films is based on photoinduced structural changes (PSC), a unique phenomenon which is found only in non-crystalline chalcogenide materials [4, 6]. The PSC in non-annealed a-As₂S₃ films (studied also in this article) are due to the photoinduced breaking of the As-As bonds by band-gap light followed by the phonon-assisted formation of As-S bonds [6, 7].

Hologram recording and self-enhancement processes in non-annealed a-As₂S₃ films are also influenced by large relaxational structural changes (RSC). RSC are inherent in all non-crystalline materials (glasses, polymers, amorphous films, etc.) due to their thermodynamic instability [8]. This instability manifests itself through internal mechanical stresses arising in the course of preparing these films (e. g. during thermal deposition in the case of a-As₂S₃ films) [2, 8, 9]. The mechanical stresses force the disordered state toward a more ordered one and diminish in the course of relaxation [2, 8, 9]. Both PSC and RSC in a-As₂S₃ films are manifested as changes in absorption and refractive indices, thickness, density, microhardness and dissolution rate [4, 9]. The changes are usually larger for PSC than for RSC; however, after holographic recording RSC can lead to significant effects such as relaxational self-enhancement of recorded holograms. RSC also influence hologram recording [10].

Self-enhancement (SE) of a non-stationary hologram is an increase in its diffraction efficiency (DE) over time under the stimulus of a single beam light irradiation or simply in the dark [2, 3]. The SE effect was reported for the first time in 1973 by Gaylord et al. [11] for phase holograms in LiNbO₃:Fe crystals. Three types of SE can be distinguished: (i) coherent SE due to the holographic recording by diffracted waves; (ii) incoherent SE due to the contrast and/or transmission increase of a hologram by incoherent light; (iii) relaxational (or dark) SE due to the contrast and/or transmission

increase of a hologram by thermostimulated relaxation processes [2, 3]. All three SE types can take place simultaneously. We have determined that in a-As₂S₃ films their relative contributions are 70, 10 and 20%, respectively. Further, if the SE process is primarily based on coherent SE as in our experiments when the SE excitation is made by one of the recording beams we also speak about coherent SE (CSE) remembering other contributions.

SE can also be regarded as a two-stage recording method, which is profitable when the recording energy or exposure time is limited at the first stage. Such a recording method also has an advantage of large vibration stability at the second stage. Recently it was used to record efficient (DE $\eta > 40\%$) phase holograms in photopolymers [12]. Using the CSE process we have also successfully recorded the image holograms in a-As₂S₃ films. SE characterises recording processes as well. It has been used by us to determine the transport characteristics and polar axis direction of LiNbO₃:Fe crystals, disorder correlation length in an amorphous semiconductor, etc. [2, 13]. SE of HG has also been reported in Bi₁₂Ti_{0.76}V_{0.24}O₂₀ crystals [14] and dichromated poly(vinylalcohol) films [15].

Does the visibility threshold – minimum $M = M_{\min}$ exist in the case of normal two-beam recording and, especially, in the case of CSE? Getting answer to this question was the motivation behind the studies reported in this article. Such a minimum visibility can exist due to the material non-homogeneity and light scattering as well as due to material non-linearity. Light scattering due to the non-homogeneity of the material takes place in all materials. In the case of a-As₂S₃ films there are some reasons to expect the effect of non-linearity on the recording modulation dependence. First, it is found that the recording light intensity threshold of about 10^{-5} W/cm² exists in a-As₂S₃ films due to RSC [16]. Photosensitivity of these films depends on light intensity [17]. The existence of minimum visibility for the recording light intensity interference pattern could explain the fact that CSE is not observed in all materials as can be expected from the CSE complementary HG model [3, 13] even when the holographic recording itself is efficient (Table 1). We have chosen an a-As₂S₃ film for these experiments because the CSE effect is the most efficient in these films.

In this article, we have experimentally studied HG recording efficiency dependence on M in both

Table 1. Existence of scalar HG coherent self-enhancement in different materials. SE factor is defined as the ratio of DE over the initial DE. λ_1 and λ_2 are the recording and readout light wavelengths, respectively.

Material	a-As ₄₀ S ₆₀	a-As ₄₀ S ₆₀	LiNbO ₃ -Fe	AC KBr	a-As ₄₀ S ₁₅ Se ₄₅ [18]	ABO T1, T2, H1, stilbene compound 11	ABO T3	Stilbene comp. 8a
Max SE factor	1000	350	50	4.5	25	<1 No CSE	1.1	42
λ_1 , nm	514.5	514.5	632.8	632.8	632.8	632.8	632.8	532
λ_2 , nm	514.5	632.8	632.8	632.8	632.8	632.8	632.8	532

normal two-beam recording case and in the CSE case. We have found the minimum recording light fringe visibility of 3.6×10^{-4} . Both HG recording and their CSE were possible down to this value. It was determined by approaching the diffracted light intensity to the scattered light background so that the diffracted signal could not be discovered any more. Theoretical explanations are provided.

2. Experiments

HG recording with a spatial period of $2 \mu\text{m}$ was made in a $7 \times 7 \text{ cm}^2$ $12 \mu\text{m}$ thick a-As₂S₃ film pro-

vided by Dr. J. Teteris from the Institute of Solid State Physics, University of Latvia. The film was thermally evaporated in vacuum at room temperature onto glass substrate. The film was not annealed. Before the experiments it had been kept in dark at room temperature for several years.

The transmission HG recording was made by two symmetrically incident Klastech DENICAFC 532–300 diode pumped solid state laser 532 nm beams, but their readout was made by the Melles Griot 25LH928-230 He-Ne gas laser 633 nm beam at the Bragg angle (Fig. 1). The lasers operated in CW mode. The recording light intensity

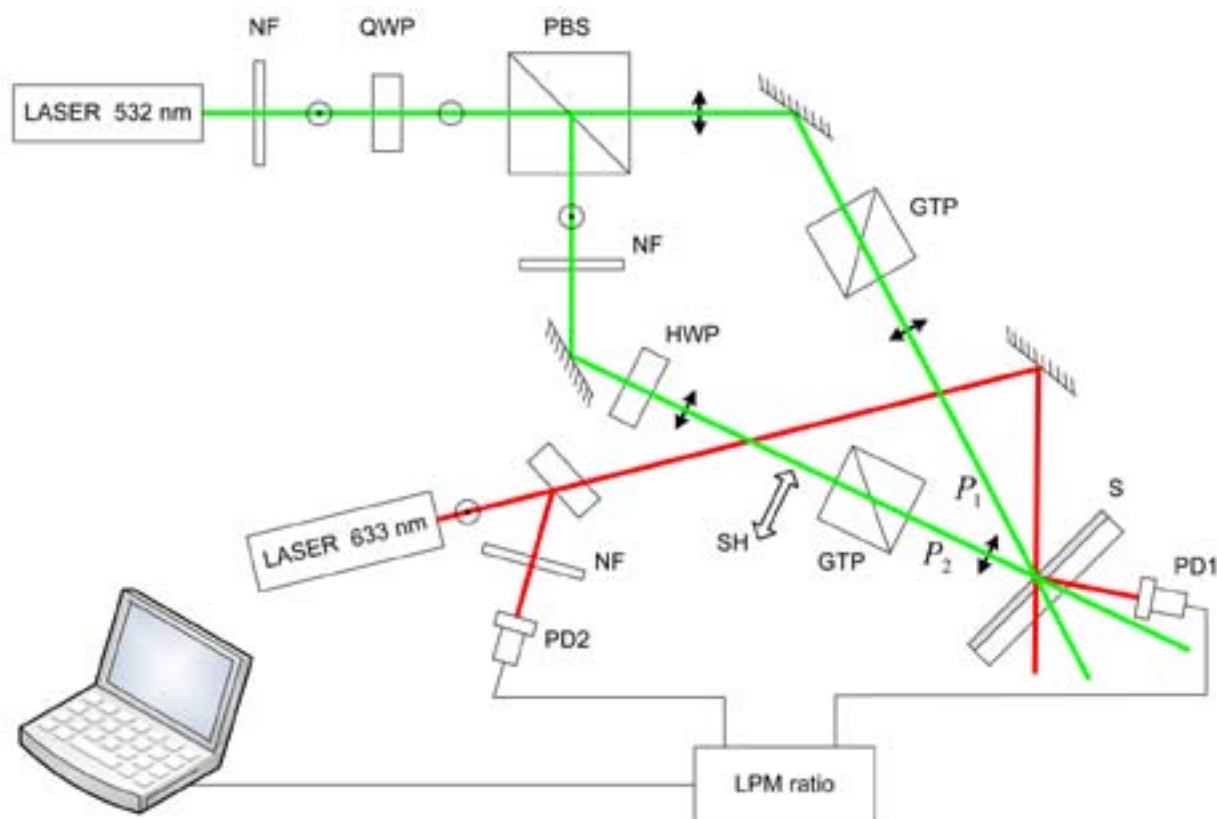


Fig. 1. Experimental set-up for HG recording and readout by two beams and by one beam in the CSE process: neutral filters *NF*, quarter-wave plate *QWP*, half-wave plate *HWP*, polarisation beam splitter *PBS*, Glan-Thompson prism *GTP*, shutter *SH*, recording beams *P*₁ and *P*₂, sample *S*, photodiodes *PD*₁ and *PD*₂, laser power meter *LPM*.

I_0 was about 0.3 W/cm^2 in all cases except for the $M = 1.0000$ case with about 0.60 W/cm^2 . This distinction was made in order to keep beam intensity exciting CSE constant ($\approx 0.3 \text{ W/cm}^2$). Fringe visibility M was varied in the $3.6 \times 10^{-4} - 1.0000$ range.

The CSE measurements were carried out as follows. First, initial HG was recorded by two beams up to a small initial DE η_i . Second, the CSE excitation was performed by the recording beam *P*₁ only (Fig. 1). The initial diffraction efficiency was kept at 0.6–0.7% (from $M = 1.0000$ to $M = 0.0324$; it was lower for $M < 0.032$), and the CSE exciting beam power was kept equal to 7.5–7.6 mW.

The first-order DE was continuously measured as the function of exposure time t by Ophir Laserstar power metre (allowing beam ratio measurements) and stored in the PC memory. DE was defined in the usual way as a ratio of the first-order diffracted beam power to the incident readout beam power. On the basis of DE measurements other quantities were determined, such as SE factor $\xi = \eta/\eta_i$, recording sensitivity $S = \eta^{0.5}/(MI_0 t)$ and CSE sensitivity $S_1 = \eta^{0.5}/(I_0 t)$. Both sensitivities are introduced according to the widespread practice [19] assuming a linear recording process. They are measured in cm^2/J . CSE sensitivity is defined without fringe visibility M , because all HG with $M \geq 0.0324$ had approximately the same initial DE 0.6–0.7% for all M but the actual visibility depends on the initial DE η_i value as will be discussed in the next section.

The recording light beam power measurement accuracy by digital Ophir Nova II laser power metre was $3 \times 10^{-4}\%$. The $1/e^2$ beam diameter of 532 nm recording beams were determined by the micrometric slit method to be $1.80 \pm 0.03 \text{ mm}$. The determination accuracy of the quantities M , I_0 , η , ξ , S , S_1 was $5 \times 10^{-4}\%$, 1.7%, 2%, 2.8%, 2.1% and 2.1%, respectively. Here it is taken into account that DE values were found from a PC screen.

The recording laser beams were *p*-polarised, readout beam was *s*-polarised. The polarisation of recording beams was chosen to minimise reflection losses. The optical elements *NF*, *QWP*, *HWP*, *PBS*, and *GTP* in the recording set-up (Fig. 1) apart from establishing the right polarisation were used to adjust the necessary recording beam powers. All the experiments were carried out at room temperature.

3. Results and discussion

The main results of our experimental studies can be formulated as follows:

(1) Scattered light background intensity is growing under exposure with one recording beam including the Bragg angle direction (Fig. 2).

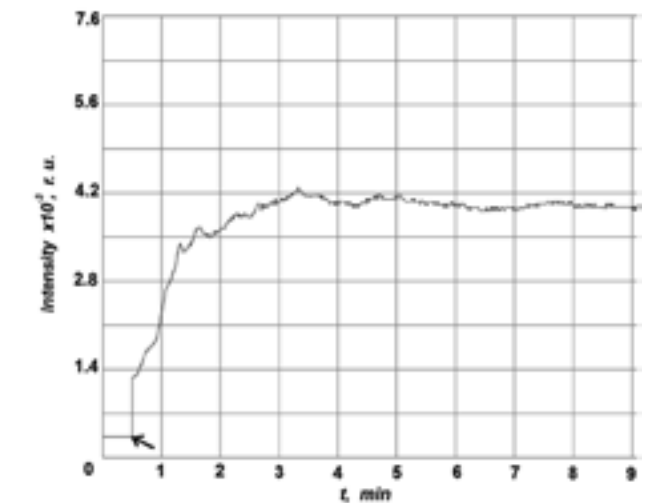


Fig. 2. Time dependence of the scattered light background intensity growth in an a-As₂S₃ film due to noise grating recording. The background light intensity was measured at the Bragg angle with one recording beam *P*₁ (Fig. 1). It is shown in relative units as a ratio of the scattered light power measured by the photodiode *PD*₁ in per cent with respect to the 633 nm readout beam power which was kept constant in all experiments. The arrow indicates the onset of the photoinduced scattering growth.

(2) In the $M = 1.0000$ case, the maximum DE $\eta_{\text{max}} = 15\%$ (Fig. 3(a)) and the maximum CSE factor $\xi_{\text{max}} = 12.3$ (Fig. 4(a)) have been found. The minimum visibility was found to be $M = 3.6 \times 10^{-4}$ corresponding to $\eta_{\text{max}} = 0.05\%$ (Fig. 3(c)) and $\xi_{\text{max}} = 5.0$ (Fig. 4(c)). This minimum modulation was determined by the fact that the diffracted light intensity reached the scattered light background.

(3) The maximal DE increased when M was increased in both two-beam recording and CSE cases (Figs. 5, 6). In contrast to this, the

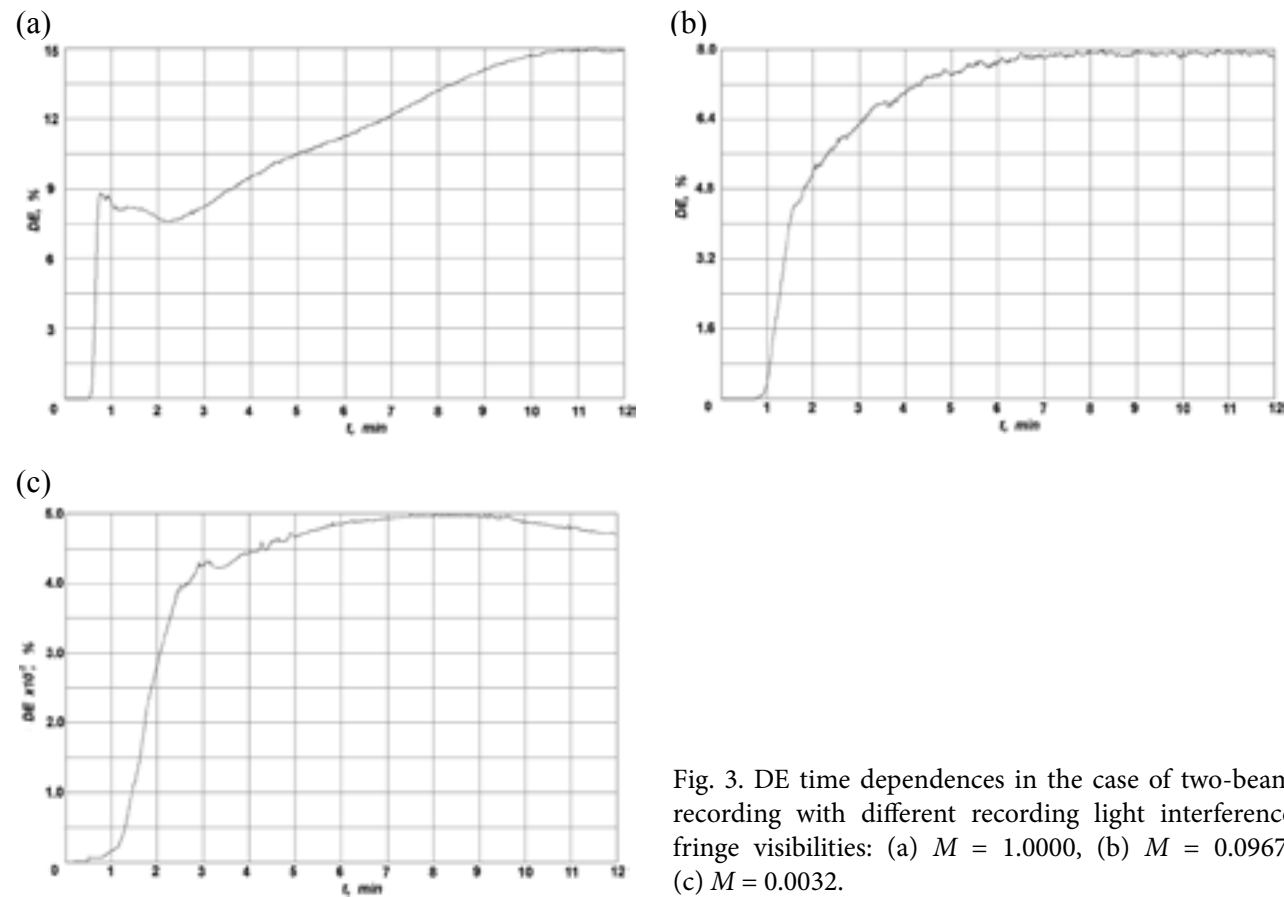


Fig. 3. DE time dependences in the case of two-beam recording with different recording light interference fringe visibilities: (a) $M = 1.0000$, (b) $M = 0.0967$, (c) $M = 0.0032$.

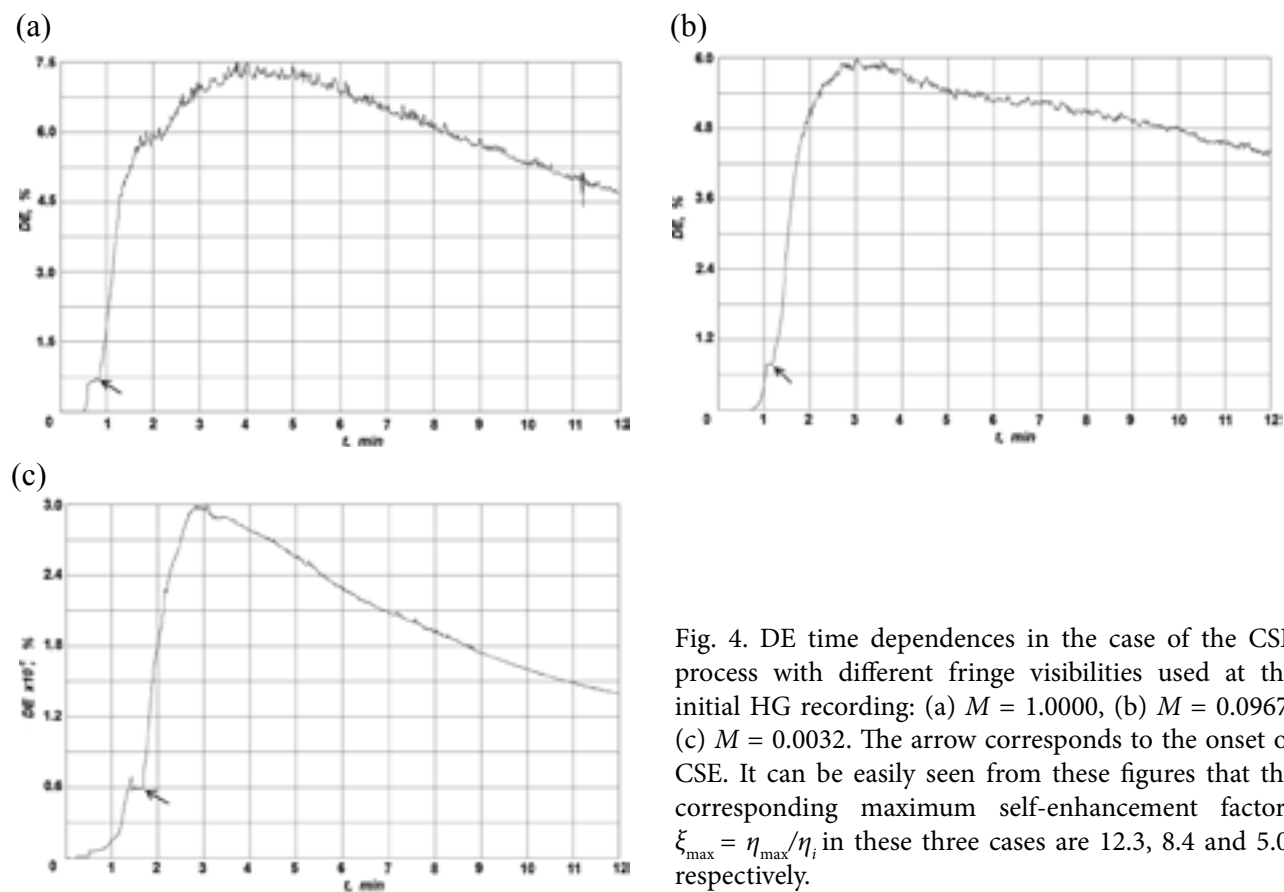


Fig. 4. DE time dependences in the case of the CSE process with different fringe visibilities used at the initial HG recording: (a) $M = 1.0000$, (b) $M = 0.0967$, (c) $M = 0.0032$. The arrow corresponds to the onset of CSE. It can be easily seen from these figures that the corresponding maximum self-enhancement factors $\xi_{\max} = \eta_{\max}/\eta_i$ in these three cases are 12.3, 8.4 and 5.0, respectively.

recording sensitivity decreased in the two-beam recording case (Fig. 5) and increased in the CSE case (Fig. 6).

Figures 3, 4 are only representative. The number of measurements was much larger. Further, the results obtained are discussed.

Non-homogeneous material can be regarded as a superposition of amplitude and phase gratings according to Fourier analysis. These gratings scatter light which records new (noise) gratings together with the incident light. This explains the growth of the scattered light background intensity even with one beam (Fig. 2).

The minimum modulation index $M_{\min} = 3.6 \times 10^{-4}$ evidently corresponds to the situation when the recorded HG amplitude becomes comparable to the amplitude of noise gratings. So the modulation threshold in a-As₂S₃ films is determined only by the background noise, but not by non-linearity of the recording. These conclusions confirm the results of a simple linear analysis of both two-wave HG recording and their CSE which is presented further.

HG in a-As₂S₃ films at 633 nm are lossy phase HG [2]. Let us assume that only photoinduced refractive index changes are responsible for the recording and that the recording is linear. The exact mathematical description of the HG recording and its CSE includes the simultaneous treatment of the wave equation and material equation. This is a rather complicated problem of non-linear optics. Instead, assuming a cosinusoidal profile of HG we shall consider only the refractive index changes in the interference pattern maxima and minima because these changes mainly determine the DE of phase HG [20]:

$$\eta = \exp(-\kappa_0 d / \cos\theta) \sin^2[\pi \Delta n_g d / (2\lambda \cos\theta)]. \quad (3)$$

In Eq. (3) κ_0 is the average absorption coefficient of HG, d is its thickness, θ is the readout beam incidence angle inside HG, Δn_g is the double amplitude of the cosinusoidal phase grating, λ is the readout light wavelength in the air. Reflection losses are neglected in Eq. (3) because they do not matter for the following analysis.

Neglecting absorption and scattering of light and taking into account Eq. (1) HG two-beam recording can be described by the following linear

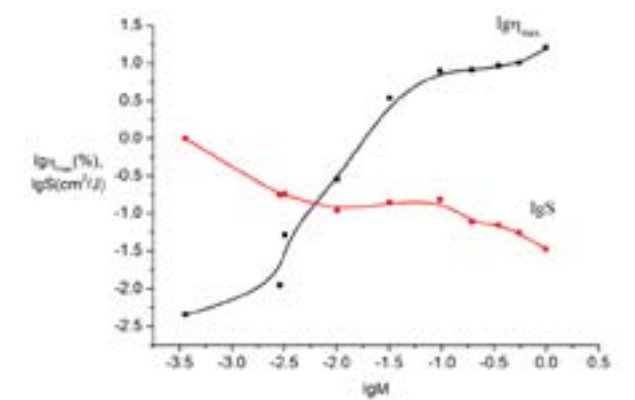


Fig. 5. Logarithms of maximal DE η_{\max} and sensitivity $S = \eta^{0.5}/(MI_0 t)$ versus the logarithm of visibility M for two-beam recording.

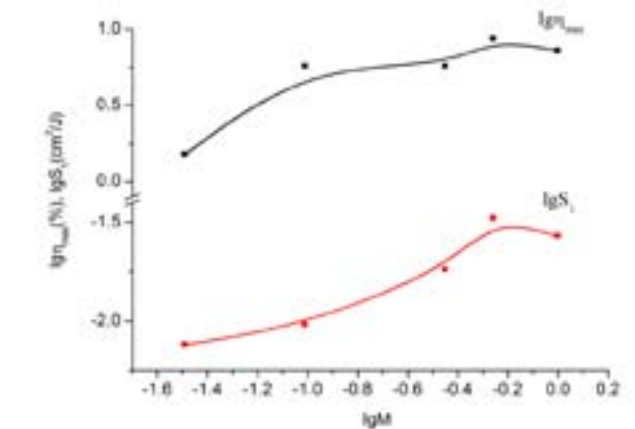


Fig. 6. Logarithms of maximal DE η_{\max} and sensitivity $S_1 = \eta^{0.5}/(I_0 t)$ versus the logarithm of visibility M for coherent self-enhancement. Initial DE $\eta_i = 0.6\%$.

differential rate equations for the interference maxima and minima, respectively:

$$\frac{d(n_{\max} - n_i)}{dt} = \gamma_{\max} (1 + M) I_0 - \frac{n_{\max} n_i}{\tau}, \quad (4)$$

$$\frac{d(n_{\min} - n_i)}{dt} = \gamma_{\min} (1 - M) I_0 - \frac{n_{\min} n_i}{\tau}. \quad (5)$$

In Eqs. (4) and (5) n_{\max} and n_{\min} are the refractive indices in the interference pattern maxima and minima, n_i is the initial refractive index,

coefficients γ_{\max} and γ_{\min} describe the photosensitivity of the material in the sites of maxima and minima assuming that photosensitivity is intensity dependent, τ is the relaxation time characterising all possible processes (population decay of excited energy levels, RSC, etc.). As mentioned, DE is determined by the double amplitude of the grating:

$$\Delta n_g = n_{\max} - n_{\min} \quad (6)$$

By subtracting Eq. (5) from Eq. (4) one gets the following equation for the HG double amplitude Δn_g :

$$\frac{d\Delta n_g}{dt} = (\gamma_{\max} - \gamma_{\min})I_0 + (\gamma_{\max} + \gamma_{\min})MI_0 - \frac{\Delta n_g}{\tau}. \quad (7)$$

The solution of this equation is:

$$\Delta n_g = [\gamma_{\max} - \gamma_{\min} + (\gamma_{\max} + \gamma_{\min})M]I_0\tau[1 - \exp(-\frac{t}{\tau})]. \quad (8)$$

Then by assuming from Eqs. (3) and (8) small enough Δn_g values at saturation one can obtain that

$$\eta_{\max} \sim \Delta n_g^2 \text{ sat} = [\gamma_{\max} - \gamma_{\min} + (\gamma_{\max} + \gamma_{\min})M]^2 I_0^2 \tau^2, \quad (9)$$

$$S \sim \frac{\Delta n_g}{MI_0 t} = \frac{\gamma_{\max} - \gamma_{\min}}{M} + \gamma_{\max} + \gamma_{\min}. \quad (10)$$

Equations (9) and (10) are in qualitative agreement with the experimental curves (Fig. 5) depicting the visibility dependence of the maximal DE and recording sensitivity. DE increase when M is increased can also be due to the non-linearity of material response [21]. As can be seen from Eq. (10), the photosensitivity intensity dependence in a-As₂S₃ films observed earlier [17] and taken into account in Eqs. (4) and (5) is a necessary condition to explain the observed S dependence on M .

The situation is much more complicated in the CSE case because a new (complementary) HG in addition to the initial one is recorded by the interference of the zeroth-order and first-order beams diffracted by the initial HG with the double ampli-

tude Δn_{gi} . The intensities of these diffracted beams are varying in time and the complementary HG is phase-shifted by $\pi/2$ with respect to initial HG [3, 13]. In this paper we shall take the simplest approach neglecting the contribution of the initial HG to the summary HG and considering only the initial CSE stage. The interference pattern visibility recording a complementary HG according to Eq. (2) is now

$$M_{CSE} = 2(\eta\eta_0)^{0.5} / (\eta + \eta_0), \quad (11)$$

where η_0 is the zeroth-order DE described by the following expression from Kogelnik's coupled wave theory [20]:

$$\eta_0 = \exp(-\kappa_0 d / \cos\theta) \cos^2[\pi\Delta n_{gi} d / (2\lambda \cos\theta)]. \quad (12)$$

From Eqs. (3, 11, 12) one gets

$$M_{CSE} = |\sin[\pi\Delta n_{gi} d / (\lambda \cos\theta)]| \approx \pi|\Delta n_{gi}|d / \lambda. \quad (13)$$

It is assumed in Eq. (13) that $\cos\theta \approx 1$ and Δn_{gi} is small enough. With the above approximations the CSE process can be treated in the same way as a two-wave recording replacing M by M_{CSE} . As seen from Eq. (13), M_{CSE} does not depend explicitly on M . Besides, the initial Δn_{gi} values in Fig. 6 were approximately the same. Thus no M dependence of η_{\max} and S_1 is expected from the point of view of our simple model. Actually, however, both quantities in Fig. 6 are increasing when M is increased.

This effect can be explained by mechanical stress modulation during the initial HG recording depending on M followed by RSC enhancing the CSE effect. More precisely, the driving force behind the relaxational SE driving force [2] is evidently larger for larger M . Thus RSC somehow "remembers" the recording interference pattern visibility. It should be noted that if sensitivity is defined as in the two-beam case (i. e. if we take S_1/M instead of S_1) then it also decreases with growing M . In terms of Eqs. (7, 8, 9, 13) with $M = M_{CSE}$ one can assume that quantities γ_{\max} , γ_{\min} and τ acquire the dependence on M due to RSC.

The developed approach may also provide an explanation why no CSE has been observed in some azobenzene oligomer films (Table 1). First, strong enough RSC as in a-As₂S₃ films can also be

a necessary condition to observe significant CSE in other (e. g. organic) amorphous films. Second, the absence of CSE can also be explained by the proposed model. It can be seen from Eq. (8) that if $|\gamma_{\max}| < |\gamma_{\min}|$, certain combinations of the parameters γ_{\max} , γ_{\min} and M_{CSE} can prevent CSE if the expression in the first brackets is zero. We have observed such unusual recording sensitivity dependence on light intensity in azobenzene oligomer [4-((4-nitrophenyl)diazenyl)-N, N-bis(2-(tetrahydro-2H-pyran-2-yloxy)ethyl)benzenamine] films [22]. However, further studies are needed to clarify these points.

4. Conclusions

Experimental and theoretical studies of the holographic grating recording and its coherent self-enhancement efficiency dependence on the interference fringe visibility are carried out in an a-As₂S₃ chalcogenide film. The minimum recording light interference fringe visibility $M_{\min} = 3.6 \times 10^{-4}$ is found, which is determined by the scattered light background. This visibility value, which as far as we know is the smallest reported value as yet, can be different when the material, recording and read-out conditions are different. The scattered light background intensity increase is observed by one recording beam due to the recording of noise gratings. In the case of two-beam holographic grating recording the maximal diffraction efficiency increases when M is increased, whereas recording sensitivity decreases. The model based on the light intensity dependence of material photosensitivity is proposed, which qualitatively explains the observed M dependence of maximal diffraction efficiency and sensitivity. In the case of coherent self-enhancement both the maximal diffraction efficiency and sensitivity increase with the initial holographic grating recording fringe visibility M in spite of the fact that actual visibility $M_{CSE} \approx \pi|\Delta n_{gi}|d / \lambda$ does not explicitly depend on M . This effect can be explained by mechanical stress modulation during the initial holographic grating recording depending on M followed by relaxational structural changes enhancing the coherent self-enhancement effect. More detailed studies are needed to clarify this effect. The developed approach may explain the absence of coherent self-enhancement in some azobenzene oligomer films.

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HOLOGRAFINIŲ GARDELIŲ ĮRAŠYMAS IR JŲ KOHERENTINIS SUSISTIPRINIMAS a- As_2S_3 PLONAJAME SLUOKSNYJE MINIMALIOS ŠVIESOS INTENSIVUMO MODULIACIJOS SĄLYGOMIS

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Siekiant nustatyti minimalų kontrastą eksperimentiškai tirta, kaip holografinių gardelių įrašymo ir jų koherentinio susistiprinimo efektyvumas a- As_2S_3 chalcogenido sluoksnyje priklauso nuo įrašomos šviesos interferencinių juostelių kontrasto. Gautas minimalus juostelių kontrastas $M_{\min} = 3,6 \times 10^{-4}$, kurį lemia išsklaidytos šviesos fonas. Šiuo atveju maksimalus difrakcijos efektyvumas $\eta_{\max} = 0,05\%$, o maksimalus susistiprinimo koeficientas $\xi_{\max} = 5,0$, lyginant su $\eta_{\max} = 15\%$ ir $\xi_{\max} = 12,3$ optimaliu $M = 1,0000$. Dvipluoščio holografinių gardelių įrašymo metu maksimalus difrakcijos efektyvumas didėja didinant M , o jautris tada mažėja. Šiems rezultatams

paaiškinti pasiūlytas paprastas modelis, pagrįstas tiesiniu įrašymu, kai erdvinį šviesos intensyvumo pasiskirstymą lemia M , ir įskaitantis nuo intensyvumo priklausančią medžiagos fotojautrią bei gardelės relaksaciją. Koherentinio susistiprinimo atveju ir maksimalus difrakcijos efektyvumas, ir jautris didėja, kai didėja juostelių kontrastas M . Tai įvyksta dėl to, kad mechaninio įtempio moduliaciją, gardelės įrašymo pradžioje priklausančią nuo M , keičia relaksaciniai sandaros pokyčiai, lemiantys koherentinio susistiprinimo reiškinį. Siūloma interpretacija gali paaiškinti, kodėl šis reiškinys nestebimas kai kuriuose azobenzeno oligomeriniuose sluoksniuose.