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DIFFRACTION IN LASER INDUCED GRATINGS ON THIN As_2S_3 FILMS

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We present an experimental study of the diffraction in laser induced phase gratings on thin amorphous As_2S_3 films. A simplified model of the energy-band structure of this material has been used to understand the light induced changes of the absorption coefficient and refractive index. The results of the diffraction experiments have been explained using the analytical predictions of the Raman-Nath and Bragg diffraction theories. The results of our study indicate a strong variation of the absorption and refractive properties of the As_2S_3 and a high efficiency of the laser induced phase gratings.

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1. Introduction

The large change of the refractive index produced by illumination with green light at the band-gap energy, the possibility to obtain samples of large size, good spatial resolution, and the possibility to erase the recorded gratings by heating, make As_2S_3 a promising material for holography, optical recording, optical phase conjugation, optical image processing, all-optical switching, integrated optics, etc. [1-5]

The investigation of the light diffraction on the holographic gratings induced in amorphous chalcogenides is important for a better understanding of the various photo-induced effects that occur in these materials and also for their applications.

We have experimentally studied the diffraction on phase gratings induced in thin amorphous As_2S_3 films. In the Raman-Nath regime (“thin” gratings), the restrictions imposed by the Bragg diffraction condition are not present, and many diffraction orders can appear in the diffraction spectrum of the reading beam. The number and intensity of the diffraction orders depend on the grating phase modulation index (related to the non-linearity of the process).

The gratings have been induced using an Ar laser (514.5 nm wavelength). At this incident wavelength, the absorption coefficient and the refractive index are both modulated in a complex, amplitude and phase grating. The light-induced change of the absorption coefficient and its temporal evolution was experimentally studied. In order to discriminate between the contribution to diffraction produced by the amplitude and the phase gratings, a He-Ne laser beam has been used for probing. In this case, the complex grating acts almost as a pure phase grating [3]. We have studied the dynamics of the phase grating, by monitoring the temporal evolution of the various diffraction orders intensities of the He-Ne reading beam.

For low modulation of the phase grating, the diffraction spectra have been explained using the analytical predictions of the Raman-Nath diffraction theory. For high modulations of the phase grating, our experimental results show an intermediary regime between Raman-Nath and Bragg, and the Bessel function approximation of the Raman-Nath model cannot give anymore an accurate prediction of the diffraction orders intensities. Our study indicates a strong variation of the absorption and refractive properties of the As_2S_3 films and the high efficiency of the laser induced phase grating.

2. Experimental results

2.1. The experimental setup

The experimental arrangement set-up for the study of the diffraction on phase gratings induced in thin amorphous As_2S_3 films is shown in Fig. 1.

We have used samples of As_2S_3 thin films (thickness, d = 5.4 μm), thermally evaporated in vacuum on glass substrate.

The linear polarized Ar^+ laser beam (λ = 514.5 nm, $h\nu$= 2.41 eV) is separated with the beam splitter BS in two beams of equal intensity which are symmetrically incident and superimposed in the As_2S_3 film. The modulation index of the grating is, in this case, m = 1. The full angle between the interfering beams (in air) is β = 13°, leading to a spatial period of the grating, Λ = 2.35 μm. For grating probing, a He-Ne beam (λ = 632.8 nm, $h\nu$ = 1.96 eV), close to the normal incidence (non-coplanar with the Ar beams) was used. The power of the diffraction orders has been measured with a silicon photo-detector, connected to a x-y recorder. The temporal evolutions of the power of the zero-th (non-diffracted light), of the first, the second and the third diffraction orders of the He-Ne reading beam were measured.

In order to ensure a good separation between the diffraction orders, the reading He-Ne laser beam was slightly tilted with respect of the plane of the self-diffraction orders of the Ar laser beams.

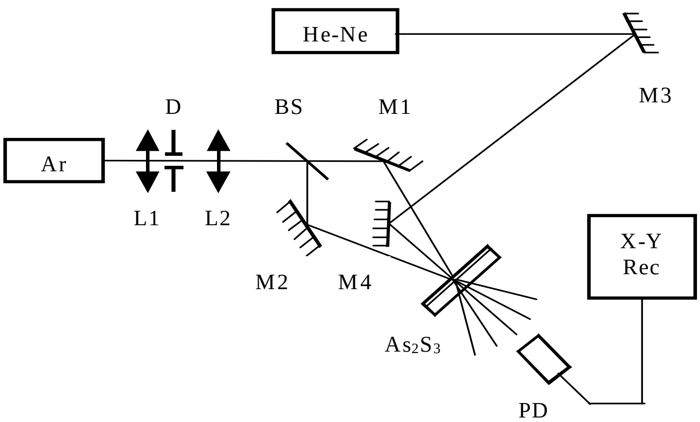


Fig. 1. The experimental setup for study of diffraction on thin As_2S_3 gratings.

2.2. Absorption measurements

We have investigate the change in the absorption coefficient, Δα, and its dynamics, produced by illumination with a single beam of the Ar^+ laser. The light-induced absorption was measured at two average intensities (I_1 = 3.5 W/cm^2 and I_2 = 0.45 W/cm^2) of the gaussian laser beam. The temporal dependence of the As_2S_3 transmission is shown in Fig. 2.

2.3. Diffraction measurements

The diffraction gratings have been recorded using two Ar^+ laser beams (λ = 514.5 nm) of equal intensity, I = 0.45 W/cm^2.

In Fig. 3 is shown the temporal evolution of He-Ne diffraction orders, recorded for different time scales: (a) 200 s, (b) 400 s, (c) 2000 s. It was obtained by writing the grating several times, in different positions on the thin film, and by recording, each time, the temporal evolution of one diffraction order of the probing beam only. The sensitivity of the measuring chain was kept constant for all diffraction orders. Some differences in the evolution of different diffraction orders and their relative magnitude, due to writing of the grating in different positions and fluctuations of Ar laser beam (the evolution of each grating was monitored for several tens of minutes), could be present.

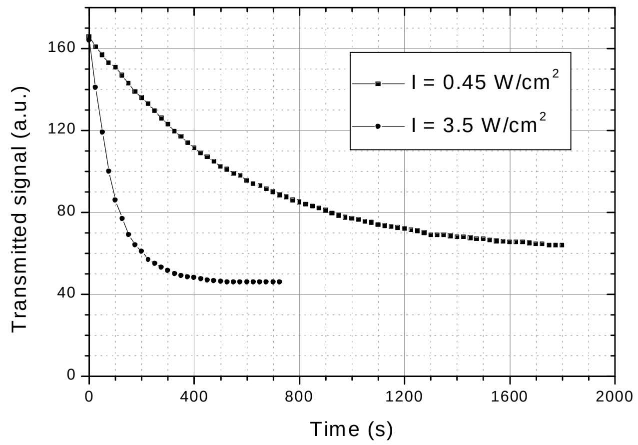
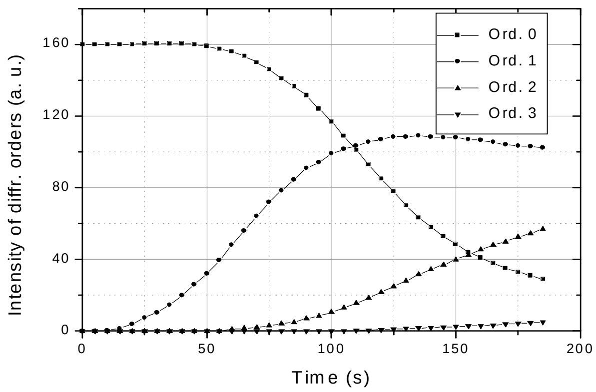
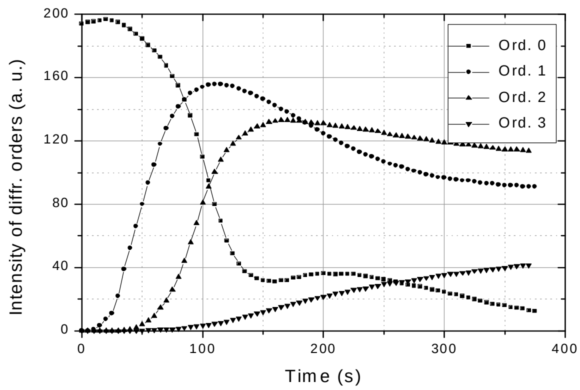


Fig. 2. The temporal dependence of the As_2S_3 transmission (λ = 514.5 nm).



(a)



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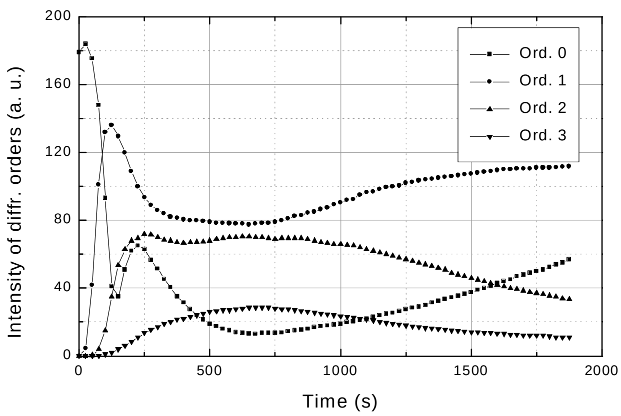


(c)

Fig. 3. The temporal evolution of He-Ne diffraction orders, recorded for different time scales: (a) 200 s, (b) 400 s, (c) 2000 s. The intensity of the writing Ar laser beams was 0.45 W/cm^2.

3. A simplified three-level model of the energy-band structure of As_2S_3 [Kwon, Kwak and Lee, [5]]

The change of the absorption coefficient and the refractive index of As_2S_3 illuminated with the green line of the Ar laser can be explained using the simplified model of the energy-band structure proposed by Kwon, Kwak and Lee [5].

The complex energy-band structure of the As_2S_3 is simplified to the energy-level diagram shown in Fig. 4.

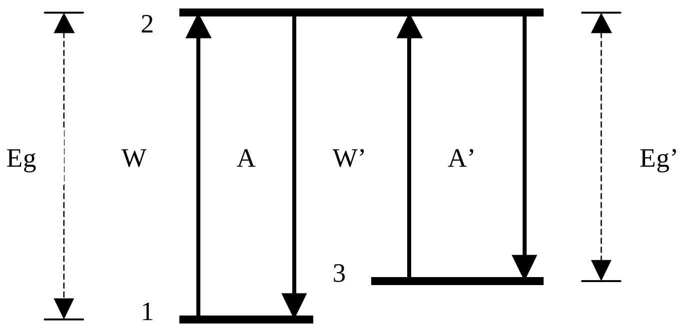


Fig. 4. A simplified model of energy-band structure of amorphous As_2S_3: level 3 - the photo-darkening state; A, A' - the spontaneous transition rates, W, W' - the pump rates.

All the valence electrons are assumed to be initially in the potential well 1. By illumination with the green line (λ = 514.5 nm, $h\nu$= 2.41 eV), valence electrons are excited to the conduction band 2 through the energy gap, E_g = 2.39 eV. Some of the excited electrons relax to the initial state 1, while others relax to the photo-darkening state 3. The pump rates 1 $\rightarrow$ 2 and 3 $\rightarrow$ 2 are W and W', respectively, while the transition rates 2 $\rightarrow$ 1 and 2 $\rightarrow$ 3 are A and A', respectively. The transition rate 3 $\rightarrow$ 1 is nearly zero at room temperature, but the transition 3 $\rightarrow$ 2 $\rightarrow$ 1 is allowed. For electrons trapped in the photo-darkening state 3, both the absorption coefficient and the refractive index are changed because of the reduced band-gap energy E'_g. Solving the rate equations for the three-level system, the temporal evolution of the absorption coefficient and the refractive index were obtained [5].

The absorption coefficient is:

$$\alpha(t) = N_1(t)\cdot\sigma + N_3(t)\cdot\sigma' = \alpha_0 + (\alpha_\infty - \alpha_0)\cdot\left[1-\exp\left(-\frac{\alpha_0}{\alpha_\infty}W't\right)\right] , \tag{1}$$

where: N_1(t), σ and N_3(t), σ' are the populations and the absorption cross sections of the states 1 and 3, respectively, α_0 and α_∞ are constant (independent of the incident intensity) and can be obtained from the experimental transmission:

$$T(t) = \exp[-\alpha(t)d] = \exp(-\alpha_0 d)\cdot\exp\left\{-\alpha_1 d\left[1-\exp\left(-\frac{\alpha_0}{\alpha_\infty}W't\right)\right]\right\} , \tag{2}$$

where $\alpha_1 = \alpha_\infty - \alpha_0$, $W' = S'I$, S' is a constant dependent on the σ', and I is the incident light intensity.

The refractive index takes a similar form:

$$n(t) = N_1(t)\cdot k + N_3(t)\cdot k' = n_0 + (n_\infty - n_0)\cdot\left[1-\exp\left(-\frac{\alpha_0}{\alpha_\infty}W't\right)\right]. \tag{3}$$

where k and k' are the contribution to the refractive index of the electrons in states 1 and 3, respectively.

4. Considerations on laser beam diffraction on phase gratings

The diffraction regime on phase gratings is determined [6,7] by the phase correlation of the diffraction orders, $Q = 2\pi\lambda d/(n_0\Lambda^2)$, and by the amplitude of the phase modulation, $\Delta\Phi = 2\pi\Delta nd/\lambda\cos\theta$ (λ is the incident light wavelength, d is the grating thickness, n_0 is the refractive index, Λ is the grating period, Δn is the modulation amplitude of the refractive index, and θ - the incidence angle of the probing beam).

In the Raman-Nath diffraction regime ($Q\leq0.5$ and $Q\Delta\Phi\leq1$, “thin” grating), many diffraction orders can appear. The intensity of the l-th diffr. order, I_l is given by [8-11]:

$$I_l = T \cdot I_{inc} \cdot J_l^2(\Delta\Phi) \tag{4}$$

where T is the transmission of the grating, I_{inc} is the intensity of the incident beam and J_l is the first kind Bessel function of the l-th order.

In the Bragg diffraction regime ($Q\geq10$ and $Q/\Delta\varphi \geq10$, “thick” grating), one diffraction order can appear only, when the Bragg condition is satisfied. Its intensity is [8-10]:

$$\theta = \theta_B : \qquad I_1 = T \cdot I_{inc} \cdot \sin^2\left(\frac{\pi\Delta nd}{\lambda\cos\theta_B}\right) = T \cdot I_{inc} \cdot \sin^2\left(\frac{\Delta\Phi_B}{2}\right) \tag{5}$$

when the reading beam is incident at the Bragg angle. When the incidence angle of the reading beam deviates with $\Delta\theta$ from the Bragg angle, the intensity of the diffraction order becomes:

$$\theta = \theta_B \pm \Delta\theta: \qquad I_1 = T \cdot I_{inc} \cdot \frac{\sin^2\left[\left((\Delta\Phi_B/2)^2 + \chi^2\right)^{\frac{1}{2}}\right]}{1+\chi^2/(\Delta\Phi_B/2)^2}\ , \tag{6}$$

where the parameter

$$\chi = \frac{\pi d \cdot \Delta\theta}{\Lambda} \tag{7}$$

is a measure of the deviation from the Bragg condition.

In the transition region between Raman-Nath and Bragg diffraction, the intensity of the diffraction orders is given by solving numerically [8,12] the set of coupled wave equations:

$$2\frac{\partial U_l(\Delta\Phi)}{\partial z} + U_{l+1}(\Delta\Phi) - U_{l-1}(\Delta\Phi) = i\rho l^2 U_l(\Delta\Phi) \tag{8}$$

where U_l is the amplitude of the beam diffracted in the l-th diffraction order and $\rho = Q/\Delta\Phi$.

5. Discussion of the experimental results

5.1. Absorption measurements

The experimental transmission of the As_2S_3 thin film (corrected for Fresnel reflections on the interfaces), was fitted with the Eq. (2), in order to obtain the pump rate W’, the values of α_0, α_∞ and, consequently, the temporal evolution of the absorption coefficient, $\alpha(t)$. The results of the fitting procedure (α_0 =10 cm^{-1}, α_∞ =2355 cm^{-1}, W’=1.75 s^{-1} for I_1 = 3.5 W/cm^2 and W’=0.21 s^{-1} for I_2 = 0.45 W/cm^2) and the corresponding curves $\alpha(t)$ are shown in Fig. 5.

5.2. Diffraction measurements

In our diffraction experiments, with the He-Ne reading beam and the experimental parameters, λ = 632.8 nm, d=5.4 μm, n_0=2.42, Λ = 2.35 μm, one can calculate Q = 1.6. In this case, the diffraction regime is the intermediary one, closer to the Raman-Nath regime. This is confirmed by the three diffraction orders that are observed in the diffraction spectrum.

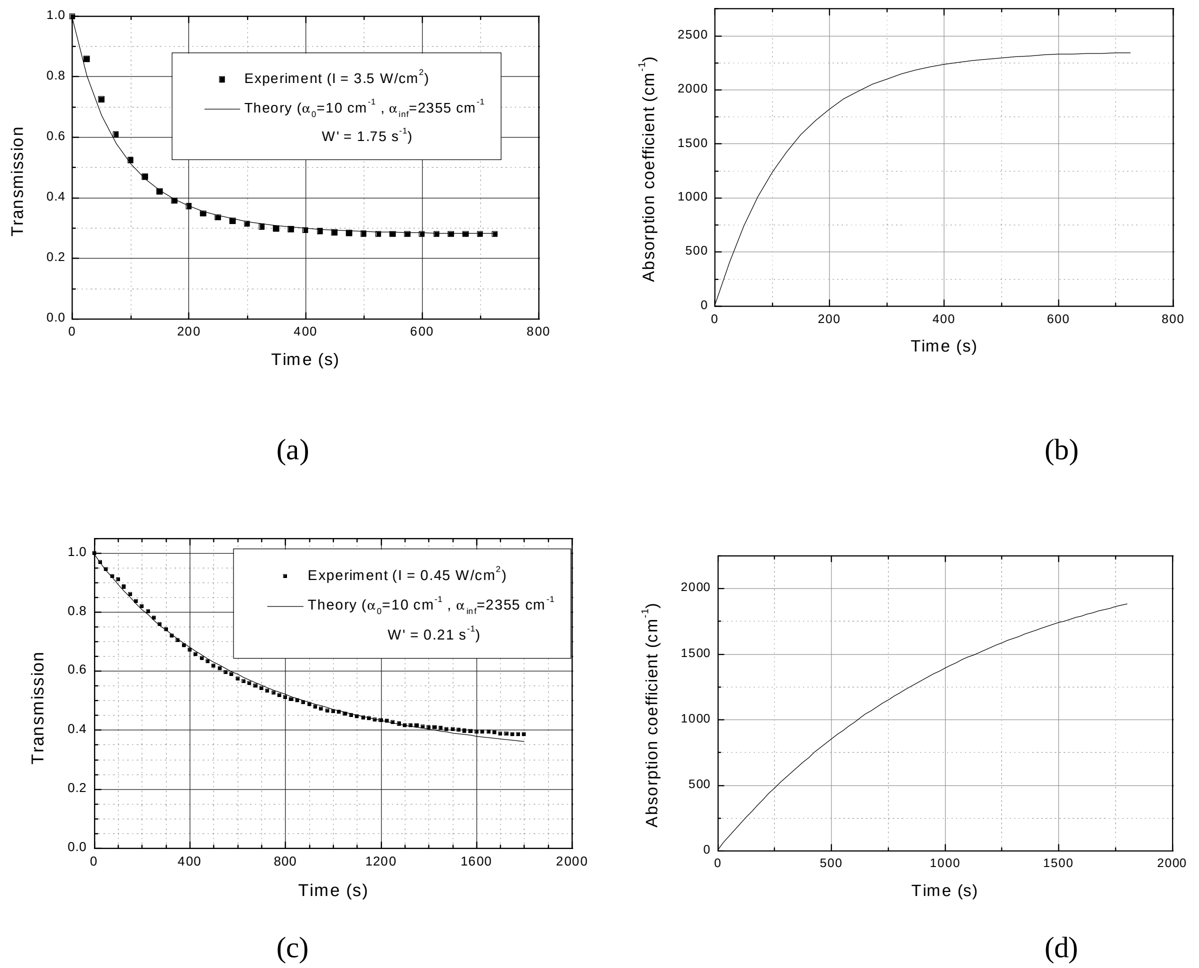
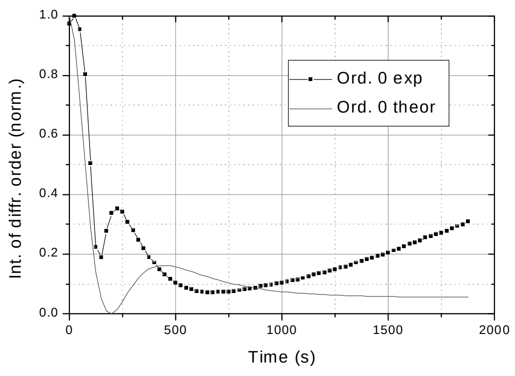


Fig. 5. The time-dependent absorption of As_2S_3 film, for I=3.5 W/cm^2 (a, b) and I=0.45 W/cm^2 (c, d).

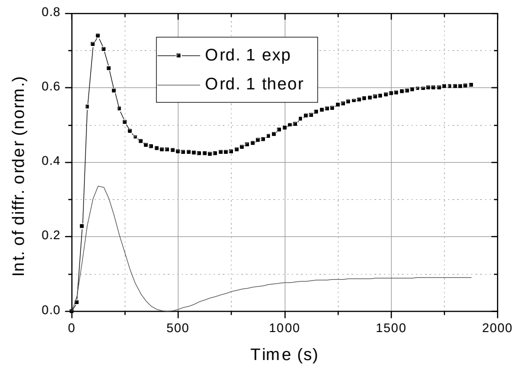
The diffraction experimental results (normalized to the value of the transmission of As_2S_3 sample, for the He-Ne laser beam) have been compared with the results of the Raman-Nath diffraction theory and using the parameters determined from the absorption measurements (α_0=10 cm^{-1}, α_∞ =2355 cm^{-1}, W'=0.21 s^{-1}). Due to the fact that the maximum intensity in the interference pattern is four times larger than the intensity of each incident writing beam, the value of the pump rate was considered W'=0.84 s^{-1}. The results of this comparison, considering n_0 = 2.42 (from literature, [13]) and n_∞ =2.51 (for a good correspondence of the temporal evolutions of the diffraction orders), are shown in Fig. 6.

The experimental results are satisfactory described by the Eq. (4) only for small refractive index modulation of the As_2S_3 grating (at the beginning of the grating recording). The discrepancy becomes large for high refractive index modulation, which could be explained by the diffraction in the intermediary regime, at the end of grating recording (when the phase modulation is $\Delta\Phi$ = 4.8, the product $Q\Delta\Phi$ = 7.7, and Eq. (4) is no longer valid).

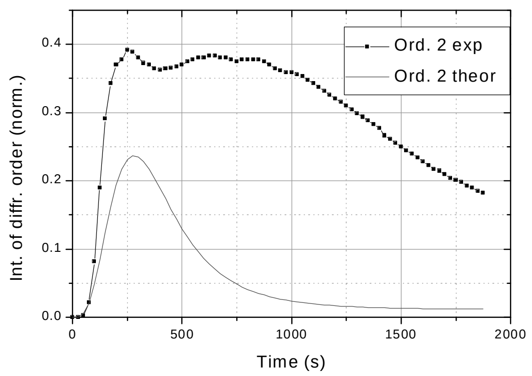
The value of the maximum diffraction efficiency in the first order, η_D = 75% (Fig. 6b), is more than two times larger than the maximum efficiency predicted by the Raman-Nath theory ($\eta_{D,\,max}$ = 33.9%), for a thin grating. Therefore, as high efficiency can be explained only by volume gratings (Bragg or intermediary diffraction regimes). The experimental results and the Bragg diffraction efficiency in the first order, considering the angular deviation $\Delta\theta$ = 4° between the angle of incidence of the probing beam and the Bragg angle (θ_B=7°45'), are shown in Fig. 7.



(a)



(b)



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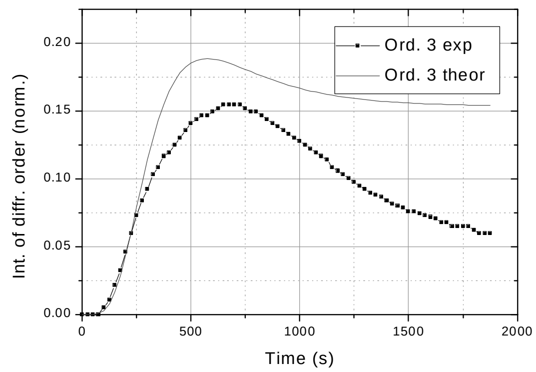

(d)

Fig. 6. Comparison between the diffraction experimental results and the Raman-Nath theory, using the temporal dependence of the refractive index predicted by the Eq. (3); W'=0.84 s^{-1}; n_∞ =2.51

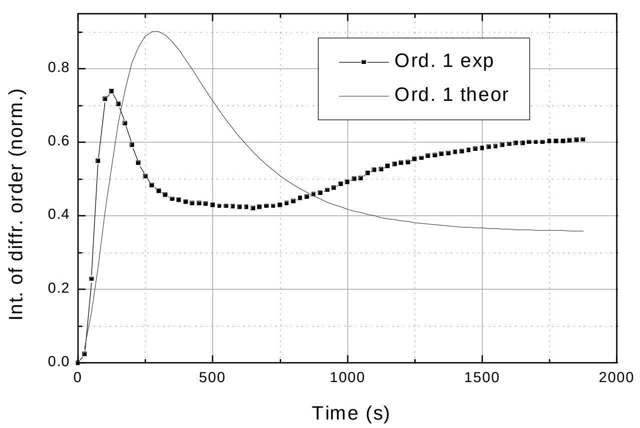

Fig. 7. The Bragg diffraction efficiency in the first order, considering the angular deviation $\Delta\theta = 4°$ between the angle of incidence of the probing beam and the Bragg angle (θ_B=7°45').

From the Figs. 6 and 7 it is clear that an accurate description of the diffraction experimental results, for the entire range of phase modulations, can be obtained only by solving numerically the set of coupled wave equations (Eq. (8)). This will be done in a future paper.

From experimental diffraction and absorption results, and using the Eq. (3), it was possible to estimate the temporal dependence of the refractive index change (Fig. 8).

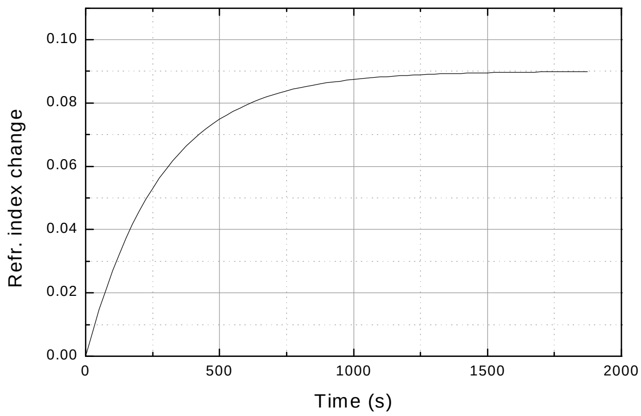

Fig. 8. The temporal dependence of the refractive index change obtained from the experimental results.

6. Conclusions

The results of an experimental study of diffraction on thin As_2S_3 gratings have been presented. The absorption of As_2S_3 thin film increases monotonically with exposure and saturates at the same value α_∞, independent of the light intensity, in the range of incident intensities used in this work (I = 0.45 – 3.5 W/cm^2). The refractive index increases monotonically with exposure and saturates at the same value n_∞, in this range of intensities. For weak exposures, the diffraction on thin gratings satisfies the Raman-Nath formalism. For large phase modulations, the diffraction takes place in an intermediary regime (between Raman-Nath and Bragg) and the diffraction efficiency can reach 75%. Our experiments can be satisfactory explained in the frame of a simplified three-level model of the energy-band structure of As_2S_3.

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