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HOLOGRAPHIC RECORDING IN NANO-SIZED As_2S_3 FILMS

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The diffraction gratings in 10, 15 and 20nm thick As_2S_3 films are holographically recorded with a totally reflected reference wave. The maximum measured values of the diffraction efficiency are 0.0005%, 0.004% and 0.007%, respectively. The exposure dependence on the diffraction efficiency is investigated. Despite of the relatively low efficiency, we succeed in the focal plane quasi-Fourier USAF test target holographic recording and reconstructed.

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1. Introduction

It is well known that chalcogenide glasses are widely used as holographic storage media [1]. For example, thick As_2S_3 films exhibit large photoinduced changes of its optical characteristics essential for the optical recording. The value of the refractive index change $\Delta n = 0.13$ has been reported in [2]. This enable achieving the diffraction efficiency value up to 80% for the holographic grating, recorded in 10μm thick As_2S_3 films [3] even in one of the first experiments [3]. In the same publication Keneman reports several percent efficiency in 2μm thick film. The reason is that the photoinduced refractive index change Δn rapidly decreases with the thickness diminishing and for 970nm thick As_2S_3 films is 0.056 [4]. On the base of spectrophotometric investigations has been established that the absorption coefficient increasing (photodarkening) disappears for thinner than 50nm As_2S_3 films [5]. On the strength of the Kramers-Kroning relations, could be expected that it is the lower limit for photoinduced changes caused by usual, plane or focused homogeneous waves.

For the rapidly developping field of nanotechnology [6] is very useful not only the application of the holographic grating measuring methods [7], but also for the nano-scale information recording. Evidently, a new optical method has to be used in order to record holographic (interference) information in nano-sized photosensitive media. The application of surface propagating evanescent waves, as it has proposed for the first time by H. Nassenstein enable the holographic recording in a very thin layer near the recording medium surface [8]. Recently, one of the authors has reported a holographic recording in 39nm thick As_2S_3 films, using a total internal reflection for the creation of the evanescent reference wave [9].

In this paper we report a further development of this method illustrated with holographic recordings in 10, 15 and 20 nm thick As_2S_3 films. The exposure dependence of the diffraction efficiency is investigated, showing maximum values of 0.0005%, 0.004% and 0.007%, respectively. The measuring are made with a He-Ne laser at 633 nm. We have recorded also quasi-Fourier hologram of the USAF test target in 20 nm thick As_2S_3 films, using a random phase mask.

2. Experimental

2.1 Sample preparation

The experiments are performed with as-deposited a- As_2S_3 films having thickness' 10, 15 and 20nm. The films are obtained by vacuum condensation of high purity As_2S_3 into planetary rotating substrates in a standard vacuum unit with an oil diffusion pump maintaining residual pressure in the order of 2 - 4.10^{-4} Pa. A resistively heated Ta crucible having a special design is used. It allowed the sublimation and evaporation of As_2S_3 without local overheating thus preventing non-desirable effects of thermal decomposition. The used substrates are pre-cleaned Ca-Na silicate glass plates. The deposition rate is 0.1nm/s at an evaporation temperature of 240^oC. The thickness of the growing films and the deposition rate are continuously measured using a quartz crystal monitor MIKI FFV. The sample thickness is additionally controlled by a surface profile recorder Talystep Rank Taylor Hobson, Code 112/1037 M-S.

2.2. Holographic recording

The holographic recording is made with the evanescent reference wave, created by the total internal reflection (TIR). This is the case, when the later approaches the reflecting interface from the denser medium with the refractive index n_1. We will consider the case of normally polarized to the incidence plane waves. If E_o is an amplitude of the falling plane wave with vacuum wavelength λ_o, the electric field amplitude falls of exponentially in TIR with the distance z into the second, rarer medium as

$$E = \frac{2E_o n_1 \cos\varphi}{\left(n_1^2 - 1\right)^{1/2}} \exp(-z/z_o) \tag{1}$$

The TIR condition is the incident angle φ to be greater than the critical angle value $\varphi_c = arcsin(1/n_1)$. In (1) z_o is the penetration depth, characterizing the decay into the second medium (air).

$$Z_o = \frac{\lambda_o}{2\pi\left(n_1^2 \sin^2\varphi - 1\right)^{1/2}} \tag{2}$$

The interference between this evanescent and object plane wave with the same amplitude is described by

$$Ie, p(x,z) = \frac{4E_o^2 n_1 \cos\varphi}{\left(n_1^2 - 1\right)^{1/2}} \exp(-z/z_o) \cos 2\pi n_1 (x \sin\varphi - z)/\lambda_o \tag{3}$$

If the recording medium thickness d is smaller than the penetration depth, as it is pointed out by Harrick [10], the TIR is not influenced by its refractive index. In this case the interference is confined near the reflecting surface, i.e.

$$Ie, p(x) = \frac{4E_o^2 n_1 \cos\varphi}{\left(n_1^2 - 1\right)^{1/2}} 2\pi n_1 (x \sin\varphi)/\lambda_o \tag{4}$$

The necessary condition is given by

$$d < z_o < \frac{\lambda_o}{2\pi\left(n_1^2 - 1\right)^{1/2}} < \frac{\lambda_o}{20} \tag{5}$$

The film thickness could be less than 25nm for grating recording with 488nm wavelength.

3. Results and discussion

The microstructure of the obtained As_2S_3 films is studied by conventional electron microscopic methods. For the purpose metal-carbon replicas from the films surface are prepared by consecutive vacuum deposition of Pt and C. The replicas observation are performed under a transmission electron microscope JEOL, JEM 100 B.

Fig. 1 shows transmission electron micrographs of Pt/C replicas from the surface of a- As_2S_3 films with thickness 10nm (a) and 20nm (b). As it is illustrated in Fig. 1(a) the As_2S_3 film is built up of individual grains separated by well -defined intergrain boundaries. Simultaneously, the halo ring of the electron diffraction is an evidence for the amorphous structure of the films. From the micrograph in Fig. 1(b) a slight increase of the mean grain size is observed for the thicker sample. These structural peculiarities of the samples studied have been already observed in a previous investigation [11].

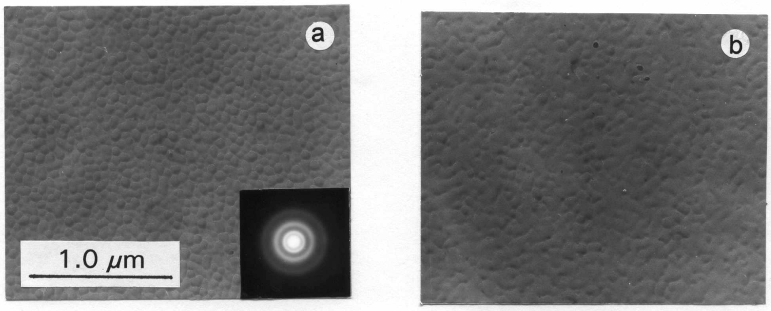

Fig. 1. Transmission electron micrograph of the samples: a-10nm thick film; b-20nm thick.

The experimental set up is sketched in Fig. 2. We have used Ar^+ laser as a light source. The initial intensity is 20mW/cm^2. For gratings recording the intensity ratio 1:1 is used in order to achieve maximum interference visibility. The object beam after the beam splitter (BS) is incidence normally on the TIR prism (P) with the refractive index 1.522. The glass substrate with the deposited As_2S_3 film is attached at the prism by microscopic oil. When the USAF resolution target is used as an object, the object beam (o) is filtered and expanded with lenses L_1 and L_2. For a noise reduction a random multilevels phase mask (Ph) is used. Reconstructed image or diffracted intensity is monitored through red filter (IRF) with CCD-camera or powermeter (PM). Unexpanded reference wave (r) is falling under incidence angle 45^o on the reflecting surface. The critical angle is 41.1^o and the penetration depth according (2) is 195nm. A He-Ne laser reconstructs the holographic recording at 633nm.

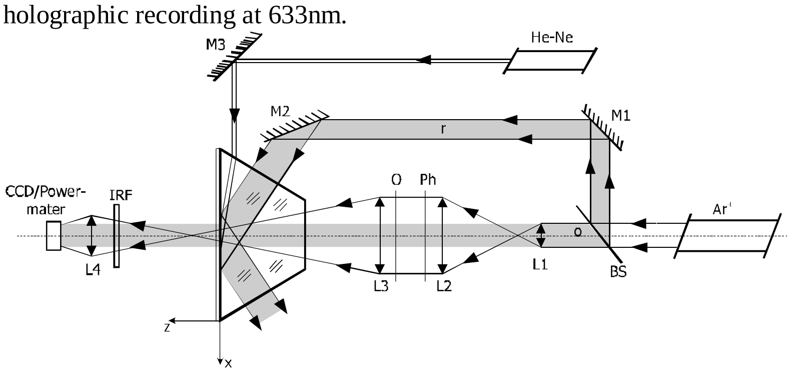

Fig. 2. Optical arrangement for evanescent-wave holographic recording: L-lences; M-mirrors; BS-beam splitter; Ph-phase mask; O-object; IRF-interference red filter.

The exposure dependence of the diffraction efficiency is shown in Fig. 3. The maximum measured values for 10, 15 and 20nm thick As_2S_3 films are 0.0005%, 0.004% and 0.007%, respectively.

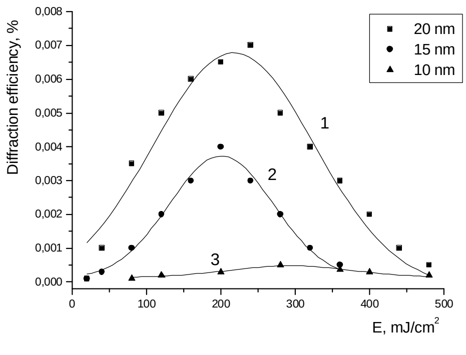

Fig. 3. Exposure dependence of the diffraction efficiency: 1-for 20nm thick As_2S_3 film; 2-for 15nm; 3-for 10nm.

After reaching maximum, the diffraction efficiency is diminishing due to overexposure like in conventional holographic case. It should be noted that optimum exposure is in the range 200-300mJ/cm^2. It is interesting to compare our results with the exposure dependence of the diffraction efficiency of ordinary holographic gratings, recorded in thin As_2S_3 films with Ar^+ laser at 488nm. To the best of our knowledge, for the smallest thickness of 350nm the optimum exposure is 7.6J/cm^2 [12]. In the first experiment, performed in 1969, this value is 3.0J/cm^2 for 450nm thick As_2S_3 films [13]. In recently published paper [14] the authors reported that the dynamic diffraction signal has maximum at 2J/cm^2 for 1100nm thick As_2S_3 film. Obviously, the optimal exposure decreases with the thickness increasing for holographic gratings, recorded with plane waves in the film's volume. On the other hand, in holographic grating recording with surface propagating reference wave, the maximum signal is obtained at lower exposure. Despite of the fact that the optical path of the monitoring red wave is about 3mm, the small photoinduced refractive index change is the main reason for the observed low efficiency. It is most likely that it is due to the adhesive forces, essential in nanoscale thickness. Relatively good signal to noise ratio, estimated roughly to be better than 20:1 enable us in recording of more complicated object. For that purpose we have used the USAF resolution target. The quasi-Fourier hologram is recorded near the focal plane with an evanescent reference wave. In Fig. 4 is shown the reconstructed image of the holographic recording in 20nm thick As_2S_3 films. A random phase mask (Ph) in the object plane is used for diminishing the influence of the recording medium limited dynamic range.

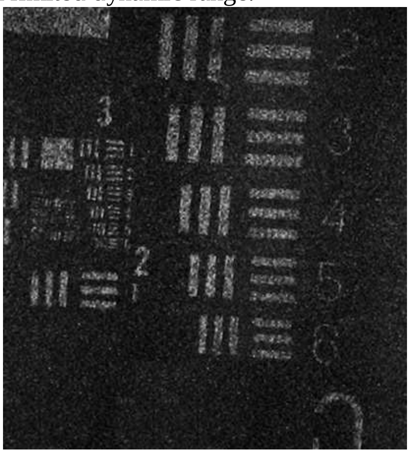

Fig. 4. Reconstructed image of the USAF target.

4. Conclusions

Using surface-propagating evanescent waves it is possible to record holograms in very thin photosensitive media with refractive index higher than input prism/substrate one. This enlarges the recording media limits for the nano-scale optical storage. Another possible application of these holograms is for grating technique that can be used as a sensitive method of investigations in nanotechnology.

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