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IMMERSION HOLOGRAPHIC RECORDING OF SUBWAVELENGTH GRATINGS IN AMORPHOUS CHALCOGENIDE THIN FILMS[♣]

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A solid immersion holographic method for the recording of refractive-index and surface-relief modulated gratings with a period of 0.2 μm – 1 μm in amorphous films of chalcogenide semiconductors As_2S_3 and As-S-Se has been developed and studied. The angular selectivity of holographic recording in amorphous chalcogenide thin films can be improved significantly by a decrease of grating period. The possibility to use the amorphous chalcogenide films as a media for holographic recording and storage of information with high density is discussed.

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1. Introduction

The most promising approach to achieving ultra-high storage density and capacity is to store data throughout the volume of a storage medium by holographic recording. It is attractive since it offers a potential volumetric density that scales with wavelength as $1/\lambda^3$, coupled with the fast parallel, page-addressed recording and readout processes [1]. It is known that the effective wavelength of light inside the media with refractive index n is reduced by a factor 1/n compared to wavelength in vacuum. Therefore an increase of storage density in high index materials can be expected.

Amorphous chalcogenide semiconductors are structurally disordered materials with high refractive index values in the range 2.2 – 4.5, depending on the film composition and light wavelength [2,3]. The photoinduced changes of refractive index down to $\Delta n \approx 0.15 - 0.5$ are observed in these systems enabling the recording of transmission holographic gratings in amorphous thin films with high diffraction efficiency [4-6]. Due to the large values of the photoinduced changes of optical properties in real time, thin films of amorphous chalcogenide semiconductors are very promising media for holographic recording, storage and processing of information, as well as for fabrication of diffractive optical elements. The photo-induced changes of wet etching rate in amorphous As-S-Se films have been studied [7,8]. Amorphous chalcogenide semiconductor (AChS) resists obtained by thermal deposition in vacuum are characterized by very high resolution capability and they possess a number of peculiarities that make them attractive for application in many photo lithographic processes [9,10].

This paper describes the application of amorphous chalcogenide semiconductor thin films in solid immersion holography being developed in our laboratory as well as describes where this method is being used and where could potentially be used.

2. Experimental details

Amorphous As_2S_3, $As_{55}Se_{45}$ and As-S-Se films with thickness of 0.1-12 μm were obtained by thermal evaporation in a vacuum of $\sim 5\times10^{-6}$ Torr onto glass substrates. The etchant, based on

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alkaline organic solution was used as a negative developer for As-S-Se films. A Ar^+ laser 514.5 nm line for recording and readout of diffraction efficiency (DE) of transmission gratings in As_2S_3 films, and a 488nm line for recording and readout of surface-relief reflection gratings in As-S-Se films were used. The holographic recording of surface-relief and refractive-index modulated gratings in amorphous films was performed by the optical schemes illustrated in Fig. 3a and 3b. The intersection of the laser beams in right angle prisms with n=1.57 -1.82 was performed.

3. Results and discussion

Diffraction-limited focusing of a laser beam for the purpose of scanning a surface to either explore or modify that surface is the basis of several important technologies. Examples include scanning optical microscopy, optical disk data recording and storage, and laser printing. The most important component of any optical focusing system is its objective lens. The size of the focused spot and the corresponding depth of focus are important factors in determining the performance characteristics of these systems.

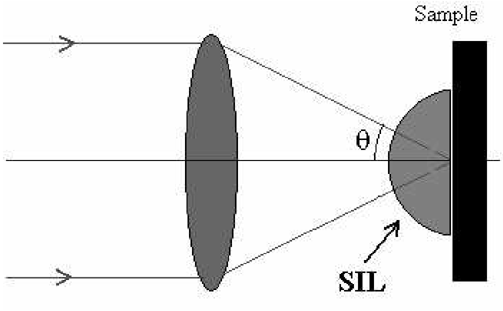


Fig. 1. Focusing through a solid immersion lens (SIL) of refractive index n_s. The effect of the SIL is to reduce the spot size by a factor of n_s.

To increase the storage density, solid immersion lenses (SILs) have been used in optical storage systems by means of increasing the numerical aperture [11]. A schematic diagram of a conventional near-field optical storage system using an SIL is shown in Fig. 1. An aberration-free objective lens focuses a collimated beam onto the SIL. All the rays pass perpendicularly through the spherical surface of the SIL without changing their propagation directions. The focal plane lies right on the planar surface of the SIL. The effective numerical aperture (NA) of the whole system can be expressed as NA= $n_s \sin\theta$, where n_s is the refractive index of the SIL and θ is a half-angle of the converging cone of light. Thus, to achieve a small focused spot, a large refractive index of the SIL is preferred. The diameter, D, of an aberration-free focused spot is given by diffraction theory as $D=K\lambda_0/NA$ or $D=K\lambda_0/ n_s \sin\theta$, where λ_0 is the vacuum wavelength of the laser beam and the value of the proportionality constant K is typically between 0.5 and 1.5, depending on the circumstances.

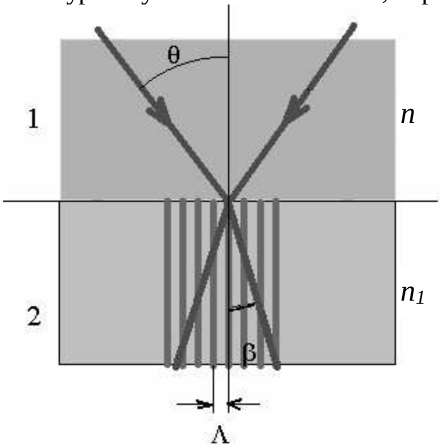
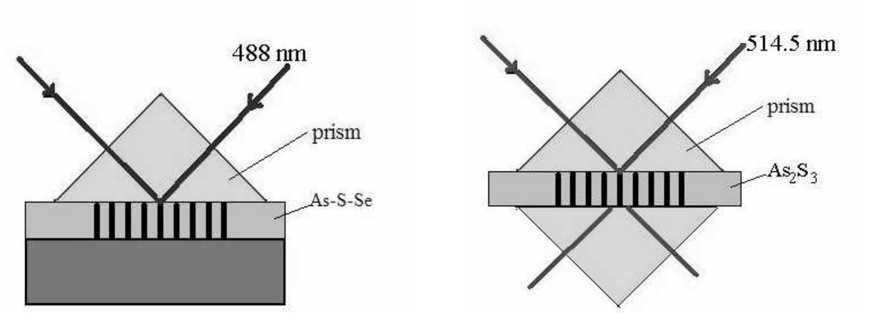


Fig. 2. Holographic recording of the interference pattern in the photosensitive material 2 with refractive index n_1 surrounded by immersion medium 1 with refractive index n.

In the case of holographic recording, when two laser beams of coherent light interfere, a pattern of parallel fringes will appear (see Fig. 2). These fringes can be used for the exposure of a photosensitive media to fabricate the surface-relief or refractive-index modulated gratings. The fringe period for two intersecting coherent light beams can be expressed as $\Lambda=\lambda_0/2\ n\ \sin\theta$, where λ_0 is the wavelength of laser light in vacuum, n is refractive index of the medium and θ is the half-angle between the laser beams in the medium. From this expression it follows that the holographic resolution of the recording material depends on its refractive index (n_1), and the smallest period that can theoretically be obtained in the recording material for $\beta=90^0$ is equal to $\lambda_0/2\ n_1$. From Snell`s law of refraction for the bending of a light ray at the interface of two media ($n \sin\theta = n_1 \sin\beta$), it is clear that to increase the angle between the laser beams inside the recording media (β), it must be surrounded by an immersion media with high refractive index (n). It can be realised by prism coupling method illustrated in Fig. 3a and 3b.



a b

Fig. 3. The optical scheme for solid immersion holographic recording of: a) surface-relief modulated gratings in As-S-Se films; b) refractive-index modulated transmission gratings in As_2S_3 films.

Amorphous As-S-Se based photoresist with refractive index n_1=3.2 at 0.488 μm were used for the recording of surface-relief gratings. The gratings were recorded by an Ar^+ laser line with λ_1=488 nm at an angle 90^o between the laser beams according to the optical scheme in Fig. 3a. After recording, wet etching of the photoresist was performed to obtain a surface-relief grating. The grating period and profile were measured by AFM. If the recording was performed in air (n=1) and the angle between the beams was equal to 90^o, a grating with a period of 0.345 μm was obtained (Fig. 4a). If the intersection of the laser beams is performed in a media with a refractive index of 1.75, using right angle prism, the obtained value of a grating period is 0.197 μm (Fig. 4b). It can be seen that the application of a prism as an immersion medium decreases the period of the recorded grating by a factor of n. The diffraction efficiency of recorded surface-relief gratings was measured using the same wavelength (λ_2= 488 nm) and prism in the Littrow mounting. The surface-relief grating in As-S-Se photoresist can be electroformed to a nickel shim and used in replication process.

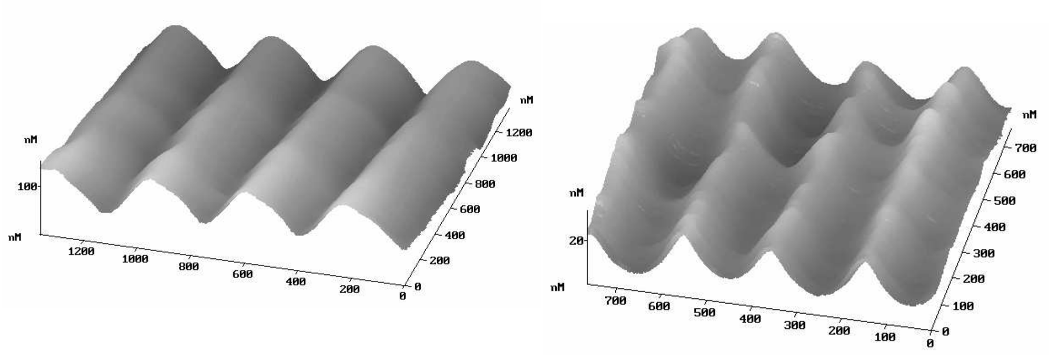


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Fig. 4. AFM picture of the surface-relief gratings recorded by Ar^+ laser line λ=0.488 μm: a) in air (n=1); a grating period Λ=0.345 μm. b) by a prism with n=1.75; a grating period Λ=0.197 μm. In both cases the half-angle between the recording laser beams is $\theta = 45^0$.

It is worth mentioning that the depth of focus of the laser interference lithography method is dependent on the coherent length of the light and can be of the order of a metre or more, compared to microns for conventional optical lithography systems. As a result, the demands on substrate flatness and resist layer positioning are not critical. Regular features with sizes of about 50 nm and less can be fabricated by this method using prisms (ZnS, GaP) with a refractive index of 3 or more as well as decreasing the recording laser wavelength. This method is perfectly suitable for production of regular patterns such as subwavelength-gratings and microsieves, as well as photonic band gap structures. It should be mentioned that such a recording method can be realized only using resists with high refractive index to avoid the total reflection by the prism-resist interface. The requirements for the resist refractive index are as follows – $n_r > n_p \sin\theta$, where n_p is a refractive index of material for the prism used and θ is the half-angle between the laser beams inside the prism. For example, if a prism with $n_p=3$ and the half-angle $\theta=60^o$ are used in the recording process, a resist with $n_r>2.6$ must be applied. The resists based on the As-S-Se system can fulfil this requirement [7].

The recording and measuring of subwavelength refractive-index modulated transmission gratings in amorphous As_2S_3 films were performed by the optical scheme illustrated in Fig.3b. The diffraction efficiency (DE) of the gratings was calculated as $\eta_n=I_d/(I_t+I_d)$, where I_d is the intensity of the diffracted beam, and I_t is the intensity of transmitted beam. The $\eta_n$enables to evaluate the real diffraction ability of the grating excluding such factors as reflection, absorption and scattering of light. The gratings up to the diffraction efficiency maximum were recorded by vertically (s-polarisation) polarised Ar+ laser λ_1=514.5 nm light beams. After the recording the DE of each grating was measured both by s- and p- polarised λ_2=635 nm light. A rapid decrease of the η_p/η_s was observed for the gratings with a period of $\Lambda<0.5$ μm [9]. Fig.5 shows the dependence of the ratio between the DEs measured at λ = 635 nm by using p- and s-polarized light on the period of the grating formed in an amorphous As_2S_3 film with a thickness of 9.3 μm. The period was changed from 0.2 to 1.0 μm. The objects behaved as thick transmission gratings. The gratings with 0.2 and 0.25-μm periods were recorded and measured by the prism coupling method. There is a satisfactory agreement between the experimental data and the fitting curve calculated starting with the cosine dependence of the angle between the readout and the diffracted light beams in the film and the n_1 value of 2.4 at the measuring wavelength of 635 nm. Consequently the subwavelength-period gratings in amorphous chalcogenide thin films possess polarization sensitive properties allowing the fabrication of highly polarization-selective elements.

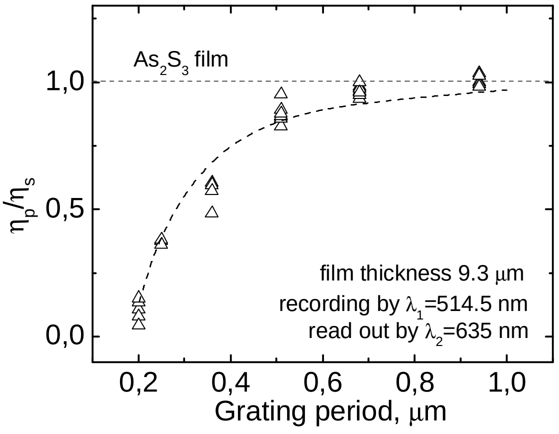

Fig. 5. Dependence of the ratio between the DE maxima, measured for p- and s-polarized 635-nm light on the period of the grating formed in an amorphous As_2S_3 film. The experimental data are fitted by a theoretical curve.

A new effective way to considerably improve the capacity and performance of current information processing and storage systems is volume storage, opening an additional dimension compared to two-dimensional, areal storage [12]. Only by using the holographic method of massive parallel data writing and retrieval on optical disks in the form of microholograms, can the speeding-up of current data-storage technology by orders of magnitude be realized. To increase the data density up to hundreds of bits/μm^2, several methods of hologram multiplexing are used, mainly wavelength and angle multiplexing.

A significant increase in angular selectivity expressed as a full width at half maximum ($\Delta\alpha_{1/2}$) of the curves representing the normalized DE vs the angle of incident was observed for the subwavelength period gratings in amorphous As_2S_3 films [13]. The linear relationship of the angular selectivity and the grating period for the fixed film thickness is illustrated in Fig. 6. It is seen that the decrease of the grating period by applying the high refractive index amorphous chalcogenide materials and the immersion holographic recording method can significantly improve the angular selectivity of holographic recording. As a result, an effective angular or wavelength multiplexing of the holographic recording in thin amorphous chalcogenide films can be realized.

The goal in any information storage system is to cram the maximum amount of data into the smallest possible space. Conventional optical surface-storage techniques are rapidly reaching their physical limitations, opening the door for volumetric approaches such as optical holography that can store data in the range of 1 Tbit on a single CD-sized disk. It is attractive since it offers a potential volumetric density that scales with the grating period as $1/\Lambda^3$, coupled with fast parallel access (potential data rates of Gbits/second order). Fig.7 shows the dependence of the maximum diffraction efficiency on film thickness and grating period of amorphous $As_{55}S_{45}$ film. The holographic recording of the gratings was performed by λ_1=632.8 nm but read-out of diffraction efficiency by 650 nm and 805 nm light. It is seen that the gratings exhibit high diffraction efficiency (η>60 %) for the films with the thickness d> 1 μm, if the read-out is performed by light wavelength λ_2=650 nm (curve 1). The diffraction efficiency does not depend on the grating period in the range of 0.246 – 0.9 μm. An essential decrease of the diffraction efficiency was observed if the read-out was performed with longer wavelength λ_2=805 nm (curve 2). These results indicate that the main factor having influence on diffraction efficiency is photoinduced change of refractive index for read-out wavelength.

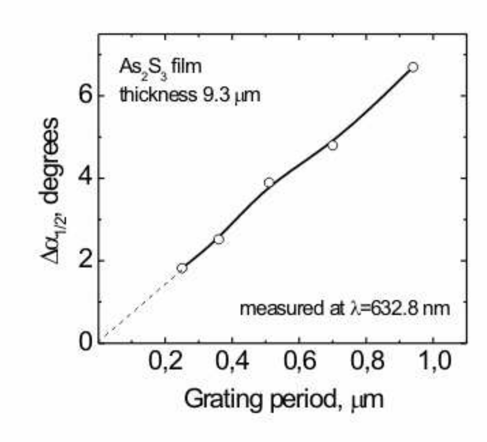

Fig. 6. Dependence of the angular selectivity on the grating period in amorphous As_2S_3 film.

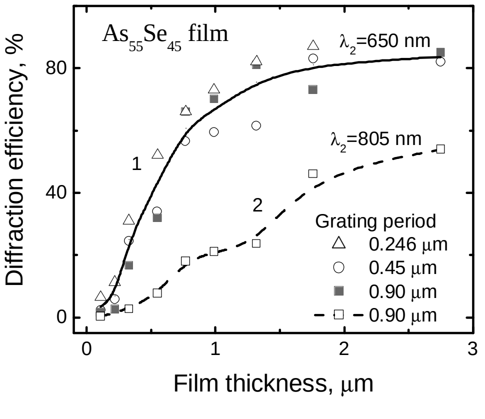

Fig. 7. Dependence of maximum diffraction efficiency of amorphous $As_{55}S_{45}$ film on the film thickness, grating period and read-out wavelength. Holographic was performed by λ_1=632.8 nm and read-out of DE by λ_2=650 nm and λ_2=805 nm light.

Storage media are central components of high-density digital data storage systems based on volume holography. The development and improvement of holographic data-storage devices causes a requirement for new light sensitive materials with high spatial resolution and in which holographic recording can be realized in real time. Using amorphous chalcogenide thin films due to their unique ability to change optical properties under laser illumination can successfully satisfy these requirements. The immersion holographic recording method is very promising in information technology. Because the effective wavelength inside the photosensitive recording material is reduced by its refractive index n_1, the amorphous films of chalcogenide semiconductors, possessing the refractive index in the range of 2.5 – 4.0, can be used as a media for holographic data recording and storage with ultra high density.

4. Conclusions

Amorphous chalcogenide As-S-Se and As_2S_3 thin films were studied as a recording media for optical holography. The subwavelength-period refractive-index and surface-relief modulated gratings with a period of 200 nm can be fabricated in amorphous chalcogenide semiconductors by immersion holographic method. It is supposed that the amorphous films of chalcogenide semiconductors, possessing the refractive index in the range of 2.5 – 4.0, are very promising media for holographic data recording and storage with ultra high density.

Acknowledgement

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