Linear Phase Shift Response with High Dynamic Range for Holographic Recording in As$_2$S$_3$

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Amorphous As$_2$S$_3$ films are real-time photographic materials for phase holograms. This work focuses on the phase shift in As$_2$S$_3$ films as a function of the exposure. A method for measuring the phase shift response and the exposure simultaneously is presented. A relative displacement determination of interferometric patterns allows precise measurements insensitive to changes of experimental conditions. The phase shift dependence on the intensity of the writing beam is evaluated for two different films. As a result, a linear phase shift response is obtained with a dynamic range of nearly $\pi$. It is independent of the intensity over four orders of magnitude. The results are proved by diffraction efficiency measurements.

Key words: interferometric phase shift measuring, holographic recording materials, linear phase recording, As$_2$S$_3$

1. Introduction

Amorphous As$_2$S$_3$ (a-As$_2$S$_3$) is a well known holographic recording material. High quality phase holograms with diffraction efficiencies up to 80% can be obtained in real-time. For that, a wavelength change is necessary between recording and reconstruction of the holograms. For writing, above band gap light ($\lambda_w=514$ nm) must be employed; for reading, below band gap light ($\lambda_w=633$ nm) must be employed. This limits the use of As$_2$S$_3$ films for volume holograms, except in the case of plane wave recording. Not restricted by the wavelength change are analog or digital 2D holograms that we use in spatial multiplexed holograms for optical vector matrix multipliers.

Despite the fact that photo-induced processes in As$_2$S$_3$ are widely studied, the holographic recording parameters of As$_2$S$_3$ films have not been investigated yet. Especially, the phase shift dependence of $\lambda_w$ on the exposure with $\lambda_w$ is important for quantitative recording of phase holograms. So this work focuses on the analysis of the phase shift as a function of exposure. Films of different thicknesses were investigated and intensities were varied over four orders of magnitude.

2. Experimental Methods

2.1 Samples

The investigated amorphous As$_2$S$_3$ films are produced by vacuum evaporation. Because of the higher light sensitivity, we use temporally annealed films that show fairly constant recording parameters after annealing for 3 months at room temperature. The same state can be achieved by annealing 10 min at 340-380 K.

2.2 Measuring Method

The wavelength change between writing and reading of holograms allows the measuring of the phase shift and the exposure simultaneously. For that, a relative interferometric measuring method was developed. This makes precise measurements possible that last several hours and are not effected by changes in the experimental conditions like room temperature and laser power. Thus the phase shift of the reading beam can be determined as a function of the exposure with an accuracy of $\pm 5^\circ$.

2.3 Experimental Setup

The complete experimental setup for the phase shift measurements is shown in Fig. 1. For writing a rectangular structure, an Ar$^+$ laser beam (single frequency mode) passes a shutter S and a beam-expanding system ($\times 5$). The intensity of the writing beam can be adjusted with a set of neutral density filters NF1. Lens L$_1$ images a rectangular slit reduced by a factor of 2.5 in the plane of the holographic film. The position of the film itself in this plane can be adjusted in the x- and z-directions. The light detectors D$_1$ and D$_2$ measure the intensity of the reflected ($I_*$) and the transmitted ($I_t$) light of the recording beam. With this setup, a rectangle of $0.4 \times 2.0$ mm$^2$ is written into the probe.

The phase-shift measurement uses the technique of a Mach-Zehnder interferometer. An He-Ne laser beam is expanded by a factor of 25 and splitted into two beams. One part of the beam is transmitted through the a-As$_2$S$_3$ film, whereas the intensity of the other part is adjusted by the neutral density filter NF$_2$ in order to maximize the contrast of the interference pattern. The plane of the a-As$_2$S$_3$ film is imaged onto the screen SC and detected by a CCD camera. The interferometer is adjusted so that a magnified image of the rectangular field that is exposed with the green light builds the major part of the interference pattern. Figure 2 shows the exposed rectangle in the interference pattern.

2.4 Data Acquisition

During the exposure of rectangles with different intensities, the transmission $I_t$ and the reflection $I_*$ of the writing beam $\lambda_w$ and the interference pattern of the reading beam $\lambda_0$ is recorded simultaneously. Two PC's equipped with an ADC board and a frame grabber board respectively are used. Each measurement consists of 200 values for $I_t$ and $I_*$ and 11 pictures of the interference pattern.

2.5 Data Processing

The measurements of $I_t$ and $I_*$ have to be normalized...
Fig. 1. Experimental setup.

Fig. 2. Interference pattern of an exposed rectangle.

and plotted after capturing. Figure 3 shows a typical plot.

The image processing of the interference pattern is more complex. Figure 2 shows a typical picture of the interference pattern. The interference stripes inside the exposed rectangle are shifted against those outside. This relative displacement is determined in the following way: 1) Profiles perpendicular to the stripes inside and outside the rectangle are plotted, 2) the period of the profile is taken as the distance of the maxima outside, 3) the displacement of the maxima inside relative to those outside is measured, and 4) the phase shift is calculated from the ratio of that displacement and the period.

Finally, the 11 consecutive phase shift measurements are plotted versus the exposure.

3. Experiments

The following parameter settings were chosen: Films with a thickness of 5.2 μm were exposed using intensities of 3.0, 1.0, 0.3, 0.1, 0.03, 0.003, 0.0003 W/cm². Films with a thickness of 1.3 μm were exposed using intensities of 3.0, 1.0, 0.3 W/cm². Except the exposures with 0.003 W/cm² and 0.0003 W/cm², all experiments were repeated three times.

Exposures with lower intensities (<0.0003 W/cm²) are of no practical use. Exposures with higher intensities were carried out, but without measuring phase shift because of the limited speed of video grabbing.

To be able to compare the results with alternative measurements, we wrote a diffraction grating (d=15 μm) with two plane waves, R and O (I_R=0.25 W/cm², I_O=0.18 W/cm²). Simultaneously to writing with λ_w=514 nm, we read the grating with λ_R=633 nm and recorded the intensities of the 0th and the first diffraction order.

4. Results

Figure 3 shows a typical plot of the reflection and transmission intensities I_r, I_t, and I_r+I_t of the writing beam (I_w=0.1 W/cm²) as a function of the exposure. The oscillations can be explained with an optical resonator inside the thin film that changes its optical length during exposure. The maxima and minima are given by the resonance condition. By this, the change of the refractive index for λ_w can be calculated by the number of maxima, the angle of incidence and the thickness d of the film. We obtained Δn_{514}=0.11 for 90 J/cm².

Apart from the oscillations, the values of I_r+I_t are reduced to 50% after exposure due to the photo-induced increase of the absorption coefficient α, which is correlated to the band gap shift to lower energies.

In Fig. 4, the phase modulation during exposure is plotted. It belongs to the same exposure as Fig. 3 (5.2-μm-thick film). The maximum phase shift is 240° at 90 J/