Effect of optical coating in the thin-film system of chalcogenide glassy semiconductor-dielectric when recording the holographic optical information

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In this paper we show that the possibility of obtaining the high values of efficiency of relief-phase holographic gratings formed in the photo-thermoplastic recording process holds not only due to the non-sinusoidal profile form, but also due to the effect of optical coating that appears in the two-layer system of photo-semiconductor – deformed thermoplastic. Analyzed are the conditions, under which takes place the antireflection optical coating, reducing unwanted reflections from surfaces and thereby enhancing efficiency. It is shown that by careful choice of the thicknesses and refractive indices of the layers materials the efficiency of holographic gratings can be improved by 2-5 %.

1. Introduction

Measuring methods of holographic interferometry are increasingly used in various fields of science and technology. Wherein are steadily increasing the requirements for accuracy and efficiency of data acquisition and analysis. It becomes commonplace when is posed the problem of creating materials and portable devices on their basis for microinterferometric measurements in real time scale. This needs creation of materials and methods with advanced optical characteristics suitable for real time registration of the holographic optical information.

Numerous studies [1-5], including ours [6-10], have shown that these requirements to the full meets the photo-thermoplastic (FTP) recording method. In the PTP method the light modulated mechanical relief is created on the surface of a thermoplastic layer. Due to the effect of "amplification" of modulation rate by the energy of electrical corona discharge field, its effective photosensitivity on some photo-semiconductors and thermoplastic layers is very high. The most high values of recording characteristics are reached on PTP recording media [7, 8, 10] based on chalcogenide glassy semiconductors of doped As-S-Se-Sn system and polymeric thermoplastic materials with optimal deformation characteristics, synthesized from different copolymers of N-vinylcarbazole group. For these media the effective photosensitivity can reach values of $\sim 10^6$-10^7 $cm^2 J^{-1}$, the resolution in some recording conditions may reach 4000 mm^{-1} [10]. Also, under those conditions the PTP carriers have the ability to form the relief-phase gratings with the diffraction efficiency (DE) approaching 40% in transmitted light. It is important to emphasize that the high values of registration characteristics can be realized simultaneously, on one and the same carrier [8]. Just satisfaction of these conditions is decisive one and permits recording holograms in a time of about 1-5 s by using the low-power laser's irradiation in green or IR domain of spectrum. Taking into account that the fabrication process of holographic gratings is quite easy and low cost, it makes PTP carriers the most suitable for real-time hologram interferometry and high-speed signal processing.

The ability of PTP process to form on the surface of thin thermoplastic films the smooth non-sinusoidal relief-phase gratings is very important, because the low cost holographic gratings with high efficiency are in great demand by various interferometric applications. The gratings with smooth, symmetric and close to sinusoidal profile may be obtained only in the case of linear response of recording media. Such gratings have theoretical limit of DE equal to 33.9% [13-15]. In case of non-linear response of the recording media (strong impact of electrical corona discharge) the grating profile is drastically different from the sinusoidal form. Experiments show [1] that in this case the first-order DE of gratings can reach 40% in transmitted light. Wherein, the efficiency is highly dependent on grating's groove shape and depth.

Below we will show, that high values of holographic gratings efficiency can be explain not only by non-sinusoidal form of their profile, but also as a consequence of optical coating arising in the thin film structure of photo-semiconductor – deformed thermoplastic. Effect of optical coating is regarded as an antireflection coating, reducing unwanted reflections from surfaces. Obtained is the assessment of the condition under which the effect will be of antireflection type.

2. Experimental

In the PTP recording method the corona discharge from high-voltage source generates homogenous distribution of positive charge on the surface of thermoplastic (Fig. 1.).
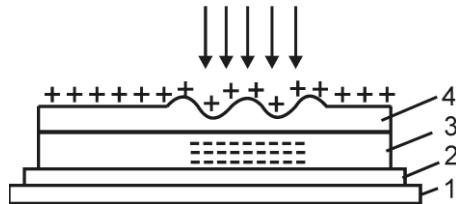
Fig.1.Photo-thermoplastic carrier: 1) flexible film, 2) transparent metal electrode,
3) CGS photo-semiconductor, 4) thermoplastic.

At hologram recording the PTP carrier is previously heated to the viscous state of the thermoplastic layer (T = 79-81ºC). Simultaneously with the exposing of a high voltage (7.2 -7.9 kV) charging unit is turned on and the thermoplastic surface is charged during 1,5 - 3 s by positive charges of the ionized air. In illuminated regions of the photo-semiconductor, an electric charge of opposed sign is induced and under the influence of the electrostatic interaction a relief-phase latent image is formed on the surface of thermoplastic layer.

PTP-media was created on a flexible polyethilenetereftalate film substrate (1, Fig.1), which was covered with the transparent chrome electrode (2). The photo-semiconductor layer (3) based on chalcogenide glassy semiconductors (CGS) of the composition of 72at.%As_2S_3:28at.%As_2Se_3 with a thickness of 1,5 μm is deposited onto the metal electrode and then is covered by a thermoplastic layer (4) based on poly-N-polyepoxypropylcarbazole with the thickness of 0,2-0,4 μm. forming electrode - semiconductor structure. For the indicated composition of CGS system the maximum of spectral photosensitivity is close to λ = 532 nm. Holographic gratings were recorded in a coherent laser radiation with the same wavelength λ = 532 nm. Diffraction efficiency of recorded gratings was defined as the ratio of light intensity in the first diffraction maximum to the light intensity passing through the unexposed area of the studied sample. This method of DE determination allows excluding the influence of light absorption in the metal electrode - semiconductor structure.

For the PTP media, the DE dependence on spatial frequency has a resonance feature while maximum DE value depends on the thermoplastic layer thickness [6]. For each PTP carrier obtained were the thermoplastic layer thicknesses corresponding to the recorded resonance frequencies near the values of 850, 900, 950, 1000 and 1200 mm^{-1}. For spatial frequencies in the region of 950 ÷ 1050 mm^{-1} the maximum values of DE were obtained near 33-37%. The narrow range of spatial frequencies is caused by two factors: by the conditions of formation of diffraction orders at a given wavelength of the laser radiation and by the magnitude of the spatial modulation *μ*

[11]. At the normal incidence of the laser beam to the surface of grating the condition of diffraction orders formation is determined by the relation: [12, 13]

$$d \sin \alpha = \frac{m\lambda}{n}, \qquad (1)$$

where *d* is the grating period, *α* – the deflection angle of the diffraction peaks, *n* – the refractive index of the medium, *m* – diffraction order, *λ* – the wavelength of the laser radiation.

From relation (1) it follows that for the laser radiation λ = 532 nm the higher diffraction orders (*m* = ± 2, ± 3 ...) will be observed at the spatial frequencies ν < 940 mm^{-1}. In this case when recording low spatial frequencies the redistribution of all intensity maximums takes place. It reduces DE in the first order of diffraction. When registering the spatial frequencies of more than 1000 mm^{-1} it is necessary to reduce the thickness of the thermoplastic layer, that leads to lower the depth of surface deformation. In this case, achieved are the lower values of the spatial modulation *μ*, which is described by the formula [12]:

$$\mu = \frac{\Delta}{d}, \qquad (2)$$

where Δ is the depth of the surface relief, d – the grating period. As shown in [4] for the relief-phase gratings maximum DE values can be achieved with *μ* > 0.4, which is impossible for thin thermoplastic layers.

To achieve the maximum values of DE, detailed studies were carried out in the range of spatial frequencies of 900 ÷ 1050 mm^{-1}. A detail study of DE dependence on spatial frequency for thermoplastic layer thickness of 0,4 μm has shown that maximum DE value of ~ 33% is reached near the spatial frequency 960 mm^{-1}. It was also found that an increasing of DE can be achieved by more accurate matching of the exposure value for each of the material compositions of semiconductor layer. Investigated were FTP carriers with different compositions of CGS and for each composition was selected radiative exposure at which the maximum DE value is achieved. The laser beams intensities were measured by the photodetector ThorLabs with an accuracy of 1 nW/cm^2. So, for the PTP carrier based on composition of 72at.%As_2S_3:28at.%As_2Se_3 and thermoplastic layer with thickness of 0,45 μm was obtained DE close to 40% at a spatial frequency of 960 mm^{-1}. From the images of surface deformation obtained by AFM method it can be seen (Fig. 2), that the thermoplastic layer is pushed through to the surface of the semiconductor layer and the depth of the deformation ~ 0,49 μm. The relief form in this case is far from the sinusoidal one.

3. Results and discussion

According to published data, the maximum diffraction efficiency for a sinusoidal phase grating may reach a value up to 33.9%, and for phase grating with a rectangular profile DE maximum value can reach up to 40.4% [11]. In case of strong impact of electrical corona discharge on

recording media (thermoplastic surface) the grating profile is smooth, but drastically different from the sinusoidal form. As can be seen from Fig. 2 in case of extremely strong impact the profile consists of periodically alternating linear and quasi-sinusoidal plots. For these profiles according to experimental data [1] the first-order DE of gratings can reach 40% in transmitted light. At this, the efficiency is highly dependent on shape and depth of profile, and thus effectively can be controlled by character of corona discharge impact.

At the same time theoretical calculations for this shape of profiles systematically provide lower values for the maximum DE. Calculations were performed on the resulting in a scalar approximation (method of spectral expansions for periodic gratings) formula for the intensity distribution in diffraction order m for non-sinusoidal gratings:

Fig. 2. AFM image and profile of the PTP carrier surface with DE ~ 40%.

$$I_m = \left(\frac{1}{N}\right)^2 \left\{ \left[\sum_{j=1}^{N} \text{sinc}\left(\frac{\beta_j d}{2N}\right) \cdot \cos\left[\alpha_j + (2j-1-N)\frac{\beta_j d}{2N} \right] \right]^2 + \right.$$

$$\left. \left[\sum_{j=1}^{N} \text{sinc}\left(\frac{\beta_j d}{2N}\right) \cdot \sin\left[\alpha_j + (2j-1-N)\frac{\beta_j d}{2N} \right] \right]^2 \right\}. \tag{3}$$

were

$$\alpha_j = h_j k_0 (n-1), \ \beta_j = (h_{j+1} - h_j) k_0 (n-1) - m\Omega, \ \Omega = 2\pi/d, \ k_0 = 2\pi/\lambda,.$$

n is the refractive index of the lattice material, d – grating period.

h_j are given by AFM measurement samples $h(x)$ defined at discrete points $j = 0, 1, …, N-1$,

N is the number of partition segments per grating's period.

Calculations by this formula give systematically underestimated by 2-5% values of DE compared with measured values. It was established experimentally that the measured closed to 40 % DE values are determined not only by the profile shape, deformation depth and the recorded resonant spatial frequency, but also depend on the processes of light reflection at the interface CGS - thermoplastic. In Figure 3a is showed an enlarged shape of the profile of two periods of grating obtained from AFM image on Figure 2. Analyzing Figure 3 we can conclude that high values of holographic gratings efficiency can be explain not only by non-sinusoidal form of their profile, but also as a consequence of optical coating arising in the thin film structure of photo-semiconductor – deformed thermoplastic. The effect of this coating can be regarded as an antireflection coating, reducing unwanted reflections from surfaces.

Let's consider the conditions under which this structure will manifest the effect of optical coating. From calculation of the thermoplastic depression and extrusion processes it can be found that relative to the initial surface
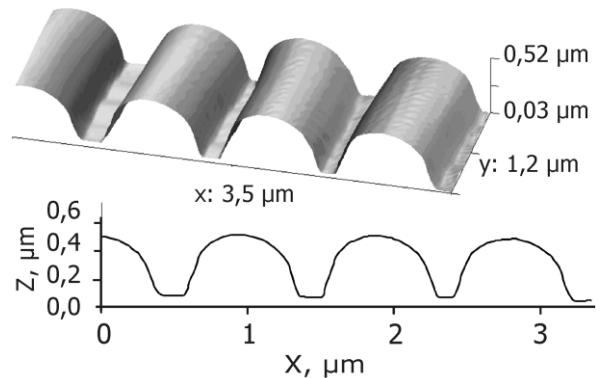
of the thermoplastic layer (dashed line Fig. 3a) surface relief has deepened on ~ 0,27 μm. The convex parts of the grooves have risen during recording on ~ 0,22 μm, so the total relief depth will be Δ ~ 0,49 μm (Fig. 3b). At the same time between the surface relief and CGS layer remains an undistorted part of thermoplastic layer with effective thickness h (Fig. 3b). The value of h can be estimated as the difference between the initial thermoplastic thickness H and the depth of punching on ~ 0,27 μm relative to the original level of the thermoplastic surface (dashed line on Fig. 3a).

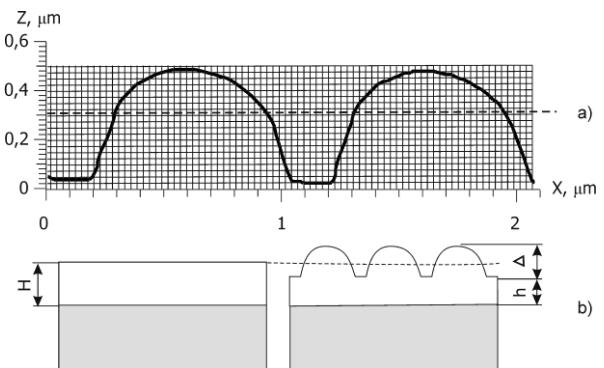*Fig. 3. Udeformed part between the surface relief and CGS layer: a) profile of grating with DE ~ 40%, b) distribution of the thermoplastic material after hologram was registered.*

At the beginning of the recording process the thickness of the thermoplastic layer was $H = 0,42$ μm. Thus, between the surface topography and layer CGS remains undistorted thermoplastic layer with thickness $h \sim 0,15$ μm.

The thin film polymer structure on the surface of CGS may have the effect of an optical coating when $h = \lambda/4$ and the refractive indices of CGS (n_{sc}) and thermoplastic (n_{tp}) satisfy the condition [12]:

$$n_{tp} = \sqrt{n_0 n_{sc}}, \qquad (4)$$

where n_0 is the refractive index of air. The measured refractive indices of CGS structure 72at.%As$_2$S$_3$:28at.%As$_2$Se$_3$ and thermoplastic material have values $n_{sc} \sim 2,6$ and $n_{tp} \sim 1,5$ respectively, which is close enough to the condition (4). In turn, the thickness of the undistorted thermoplastic layer h is estimated as $h \sim 0,15$ μm, which is close to one-quarter of the wavelength of the light ($\lambda = 532$ nm). Thus, all the standard requirements for an optical coating layer are met: the undistorted thermoplastic layer is a quarter-wave layer with refractive index between air and CGS material. It is known that such coatings can reduce the reflection for ordinary glass from about 2% to 4% per surface unit [12].

To confirm and specify numerical data on the impact of optical coating in thin film structure of thermoplastic – CGS, effect of coating has been investigated experimentally. On the CGS surface of composition 72at.%As$_2$S$_3$:28at.%As$_2$Se$_3$ was deposited a layer of thermoplastic material with thickness of $\sim 0,15$ μm and measured was spectral transmittance of CGS layer without thermoplastic one (curve 1, Fig. 4) and of the structure CGS - polymer (curve 2, Fig. 4.).
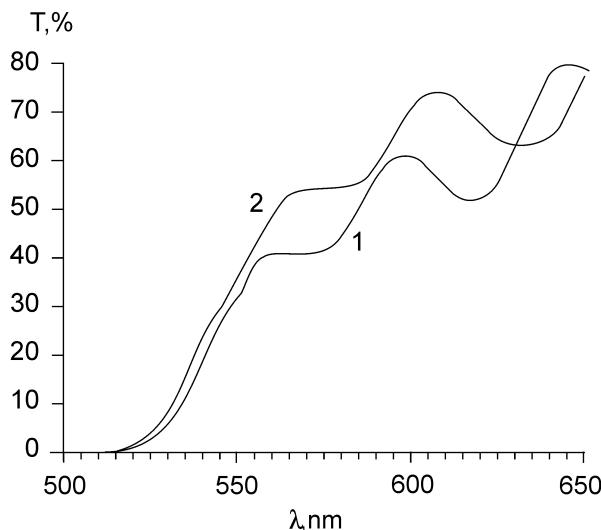

Fig. 4. The spectral dependence of transmittance:1) through a layer of CGS, 2) through the structure of the CGS - thermoplastic.

As can be seen from the spectral dependence, the transmittance of structure CGS - polymer at the wavelength $\lambda = 532$ nm is 1.18 times greater than the

transmission of CGS layer not coated with thermoplastic. From this result we can conclude that close to the limiting 40% high DE values for photo-thermoplastic carriers are determined not only by the shape profile of gratings, but also by the effect of optical coating of CGS layer by a thermoplastic one.

4. Conclusion

It is shown that close to $\sim 40\%$ limit of holographic gratings efficiency value can be explain not only by non-sinusoidal form of symmetric grating profile (grooves shape and depth), but also by impact of optical coating of photo-sensitive layer by the undistorted part of thermoplastic layer. The undistorted part of thermoplastic layer can be regarded as an antireflection coating, reducing unwanted reflections from surfaces.

A numerical evaluation has been performed and was showed that the effect of the optical coating allows you to raise the diffraction efficiency of the recorded gratings up to 40% in the transmitted light.

Acknowledgments

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