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WIDE SPATIAL FREQUENCY EVANESCENT WAVE HOLOGRAPHIC RECORDING IN PHOTOPOLYMER

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Holographic recording with referent evanescent wave and plane or evanescent object wave in photopolymer is reported. Total internal reflection is used for the creation of the evanescent waves. The spatial frequency of the recorded permanent grating vary between 150 and 5200 lines/mm. A He – Ne laser at 632.8 nm and heavy flint – glass hemicylinder are used in recording. The dependence of diffraction efficiency on the exposure is investigated.

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Ke words: Holography, Evanescent waves, Photopolymers

1. Introduction

Photopolymers are very attractive optical recording media with real - time image development, wide spectral sensitivity, long live times, good optical properties, etc [1]. Numerous investigations are devoted not only to confirm that characteristics, but also to elucidate the mechanism of holographic grating formation in photopolymers. Very thorough review of this problem is recently made [2], where non - local diffusion model, proposed by J. Sheridian [3] is further developed. It is essential to emphasize that all theoretical investigations are based on one - dimensional diffusion model, but, in fact, the holographic gratings are recorded in formulation layers thicker than ten or more micrometers. Another interesting problem is the diffraction efficiency (DE) behaviour at high spatial frequencies. To the best of our knowledge, first Colburn and Haines [4] are examined the exposure characteristics and spatial frequency response in relatively wide range -33 - 2000 lines/mm of Dupont photopolymer. The objectives of the present experimental work is not only to investigate these characteristics at higher spatial frequencies, but also holographic recording in very thin photopolymer layers. In order to record such gratings we have used the method of the evanescent wave hologram (EWH), proposed more than thirty years ago [5,6]. Recently this evanescent wave technique of lithography has been developed in our group [7,8]. In this photopolymerization process, the photoresist solidifies only in the evanescent area. It is particularly interesting for the fabrication of very thin films - thinner than 100 nm with good mechanical stability and optical transparency. The photopolymerizable formulation (pentaerythritol triacrylate + eosin + methyldiethanolamine) have refractive index change from 1.49 to 1.52.

This is the first report of a wide spatial frequency holographic recording with referent evanescent wave. By changing the incidence angle of the object beam we have recorded EWH grating with 150, 570, 1540, 2700, 3860 and 5200 lines/mm. The diffraction efficiency of 0.12 % at 150 lines/ mm falls down after 1000 lines/mm to constant values 0.025%.

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2. Experimental

2.1. Recording Medium Preparation

The present photopolymerizable system is similar to that used before [9]. The difference is in replacing a sensitizer dye -Eosin Y (2', 4', 5', 7' - tetrabromo-fluorescein disodiumsalt) with methylene blue, that has two absorption bands – at 612 and 664nm. Other components are the same -an amine cosynergist - methyldiethanolamine (MDEA) and a multifunctional acrylate monomer - pentaerythritol triacrylate (PETIA). The refractive index of non-illuminated photopolymer at 632.8 nm, measured by the proposed in [10] method is 1.488 ± 0.001 . The refractive indices of the pentaerythritol triacrylate and methyl dietholamine are 1.484 ± 0.001 and 1.466 ± 0.001 , correspondingly. The reflection spectra of the formulation, made with Carry 5E spectrophotometer with the accuracu of 0.5% [11], is shown in Fig.1.



Fig. 1. Reflection spectra of the formulation.



Fig. 2. Experimental setup: L - laser; M - mirrors; BS - beam splitter; PM- power meter; P - prism; RM- recording medium.

2.2. Evanescent - wave holographic recording

The experimental set-up is shown in Fig. 2. A 5 mW He – Ne laser "Melles Griot" emitting at 633 nm is used as the actinic source. After the beam splitter (BS) the referent beam has a constant angle of incidence - 80 deg on the hemicylindrical prism. It is made from TF – 4 heavy flint glass with refractive index $n_{1.}$ = 1.735. The spatial frequencies of the recorded grating are varied from 150 to 5200 lines/mm by moving mirrors M ₂ and M ₃ In this study we have used TE – polarized waves for holographic recording.

The spatial frequency of evanescent – wave gratings v is dependent not only on the incident angles of reference (φ_r) and object wave (φ_o), but also on the refractive index of hemicylinder, serving as an optically denser medium:

$$\nu = \frac{n_1(\sin\varphi_r \pm \sin\varphi_o)}{\lambda_0} \tag{1}$$

where λ_0 is a laser wavelength; φ_i are incident angles; n_1 is a refractive index of hemicylinder.

The evanescent wave is created by the weakly attenuated total internal reflection (TIR) from the boundary between flint – glass face and formulation. The total reflection condition is determined .by a relative refractive index (n) that is a ratio between formulation s refractive index and n_1 The TIR takes place when

$$\sin \varphi_i > n \tag{2}$$

The penetration depth of the reference evanescent wave is:

$$Z_{o} = \frac{\lambda_{0}}{2\pi n_{1} \left(\sin^{2} \varphi_{r} - n^{2}\right)^{\frac{1}{2}}}$$
(3)

When the grating is recorded with two evanescent waves, the resulting penetration depth is about half of this value. In present experiment Z_0 is 120nm.

In our experiment the formulation is spread on the substrate, made also of the TF - 4 heavy flint glass and after that this plate is attached to the hemicilinder reflecting face. A high refractive index liquid - diodmethan $CH_2 I_2$ is used for obtaining a reliable optical contact. The refractive index at 632.8 nm is 1.731. The small differences of the refractive indices between the prism's and $CH_2 I_2$ minimized the Fresnel's losses.

The recording set-up is mounted on the "Newport" PS 2000 vibration-isolating table. The recording procedure constitutes the following successive stages:



Fig. 3. Diffraction efficiency dependence on different spatial frequency

- 1. Uniform spread of 1 2 drops of the photopolymer on the substrate with Pasteur pipette for obtaining ~ $20 \,\mu$ m thick layer.
- 2. Attaching the obtained ready for recording plate with CH₂ I₂ to thehemicylinder's reflecting face.
- 3. Pre exposure 0.5s illumination with 1.0 mW/cm^2 reference beam.
- 4. Washing the exposed plate with ethanol for cleaning and removing of a non-exposed photopolymer.

After these procedures the diffraction efficiency is measured with the reference He-Ne laser beam and a « Coherent » power meter, model 212. In Fig. 3 a) is demonstrated the exposure dependence of the DE of relatively low spatial frequency gratings – 150, 570 and 1540 lines/mm. The same dependence of higher spatial frequencies – 2700, 3860 and 5200 lines/mm is shown in Fig. 3 b). The solid lines represent the best fit to the measurements at different exposures.

3. Results and discussion

In this research the holographic gratings are recorded in wide spatial frequency region. It could be mentioned, that in this case, analogous to recording in thick photopolymer layer [4], the abrupt spatial frequency response fall is observed after 1000 lines/mm. It is illustrated in Figure 4, where the normalised DE dependence on spatial frequency is shown. After 1500 lines/mm the measured DE is constant -0.02%. This value, in the limits of the measurement error, is the same, measured and recorded at 6380 lines/mm with 514.5nm wavelength [9]. It is interesting to note that only grating with lowest spatial frequency (150 lines/mm) has a DE maximum at $3J/cm^2$ exposure. In very wide spatial frequency range 570 - 5200 lines/mm the optimal exposure is almost constant $-1J/cm^2$. One possible explanation is connected with the spatial mass transfer, taking place in the holographic recording [12]. In this investigation it is predicted the decrease of the recorded efficiency at lower spatial frequencies. It could be supposed that more illumination is needed when interference fringes are far from each other.



Fig. 4. Normalised DE dependence on spatial frequency.

Despite the relatively low DE, good signal - to - noise ratio (better than 100:1) of the evanescent - wave holographic recording make photopolymers attractive for the lithography.

Finally, we believed that the results of a real one – dimensional recording will help the theoreticians to elucidate the mechanism of holographic grating formation in photopolymers.

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References

- H. J. Coufal, D. Psaltis, G. T. Sincerbox (Eds.) "Holographic Data Storage", p. 171, Springer – Verlag, Berlin, 2000.
- [2] S. D. Wu, E. N. Glystis, J. Opt. Soc. Am. B, 20, 1177 (2003).
- [3] J. Sheridian, J. R. Lawrence, J. Opt. Soc. Am. A 17, 1108 (2000).
- [4] W. S. Colburn, K. A. Haines, Appl. Opt. 10, 1636 (1971).
- [5] H. Nassenstein, Phys. Letters 28A, 249 (1968).
- [6] O. Bryngdahl, J. Opt. Soc. Am. 59, 1645 (1969).

- [7] C. Ecoffet, A. Espanet, D. J. Lougnot, Adv. Mater. 10, 411 (1998).
- [8] A. Espanet, C. Ecoffet, D. J. Lougnot, J. Polym. Sci. A. 37, 2075 (1999).
- [9] S. Sainov, A. Espanet, C. Ecoffet, D. J. Lougnot, J. Opt. A: Pure Appl. Opt. 5, 142 (2003).
- [10] S. Sainov, N. Dushkina, Appl. Opt. 29, 1406 (1990).
- [11] I. Konstantinov, T. Babeva, S. Kitova, Appl. Opt. 37, 4260 (1998).
- [12] G. M. Karpov, V. V. Obukhovsky, T. N. Smirnova, V. V. Lemeshko, Opt. Communs. 174, 391 (2000).