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Multiplexed holograms in phenanthrenequinone–polymethylmethacrylate composite for microscopic applications

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Abstract: The conditions of multiplexed hologram recording in the layers of polymethylmethacrylate (PMMA) with a high content of photosensitive components, phenanthrenequinone (PQ), were analyzed theoretically and experimentally. The sequential record of up to 10 holograms onto a single photosensitive layer with a possibility of angular selection of holograms by their reconstruction was realized experimentally. The process of the alignment of values of the diffraction efficiency by selecting of exposing dose at each sequential hologram record was shown. The possibility to use multiplex holograms based on PQ-PMMA layers in application to 3D-microscopy was demonstrated.

OCIS codes: (090.0090) Holography; (050.7330) Volume gratings; (190.7070) Two-wave mixing.

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1. Introduction

The use of a Volume Holographic Imaging System (VHIS) in a microscope attracts rising attention [1,2]. Usually mechanical scanning or refocusing is required in microscopes to map

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different depths within an object. To this aim, we use volume-phase-holograms (Bragg) possessing a multiplexing feature. Multiplexing means that several volume holograms are superimposed (multiplexed) on the same area of the recording material. This allows us to simultaneously project several wave-fronts originating from particular depths within the sample on a single imaging plane [2]. For good performance not only a high but also a uniform diffraction efficiency (η) is required. Yet, upon their sequential recording, η tends to decrease with increasing number of stored holograms [3,4]. This closely relates to the detailed mechanism of grating formation in the holographic material, still allowing moderately high diffraction efficiencies. Five multiplexed grating with unequal η between 17% to 46% could be achieved in [1] using pre-illumination and varying the dark delay time between exposures.

Phenanthrenequinone - polymethylmethacrylate (PQ-PMMA) composite is a widely used polymer for holographic recording [5–7]. These highly-concentrated PQ-PMMA layers with 100 µm thickness have demonstrated the effective recording of transmission and reflection holograms [8,9] as well as the formation of the waveguiding structures [10] exposing with an Argon laser (at 488 and 514.5 nm). Recently we discussed in details the formation of stable phase reflection holograms in highly-concentrated PQ-PMMA layers [11]. Three main stages were included: 1) holographic recording by exposing with the interfering light; 2) thermal treatment to increase the diffraction efficiency; and 3) uniform light treatment, to make the hologram insensitive to further light exposure.

In continuation, we present here the recording of angular multiplexed volume phase holograms in PQ-PMMA to generate images from different depths of the sample. We also discuss the optimization of multiplexing process.

2. Material and experimental techniques

Holographic layers (100 µm thick) were prepared by pouring the liquid solution of PMMA and PQ (3 mol.%) on a glass substrate with subsequent drying. To analyze an ability of the material to record angular multiplexed holograms the Denisyuk experimental set-up was used as described in [11]. Diffracted intensity and angular selectivity curves were measured at λ = 532 nm laser irradiation, which is in the absorption band of PQ-PMMA material (from UV up to 540 nm [11]).

3. Holographic multiplexing

Multiplexed thick reflection holograms in PQ-PMMA composite were recently reported [11]. For a relatively wide range of the recording angles (5 ÷ 15°) an angular selectivity better than 1° could be shown for each of the multiplexed holograms. Using Kogelnik's formulas [12], angular selectivity curves were calculated by assuming that several reflection holograms were recorded by light incident from different angles but at one point of the material layer (Fig. 1(a)–1(c)). The diffraction efficiencies, assumed equal, were calculated as [12]:

$$\eta = \tan^2\left(\frac{\pi l}{\lambda \cos\theta_0}\cdot\frac{\Delta n}{N}\right), \tag{1}$$

with the layer thickness l, the Bragg angle θ_0 measured in the medium, the probing wavelength λ measured in the air, the total number of exposed holograms N and the modulation amplitude for the refractive index of a single hologram Δn (typical value: 10^{-4}). This predicted that well separated hologram performance can be expected, which was confirmed by our corresponding experimental data (see Fig. 1(d)–1(f)).

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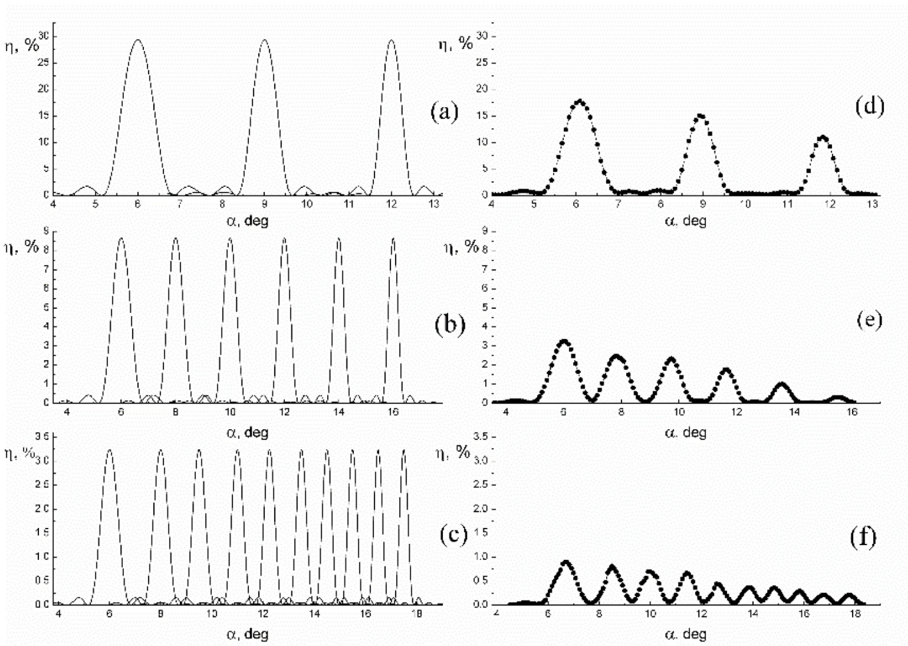

Fig. 1. (a–c) Theoretical and (d–f) experimental curves of angular selectivity for 3 [a, d], 6 [b, e] and 10 [c, f] angular-multiplexed holograms.

Nevertheless, the diffraction efficiencies of the recorded holograms appeared to be less than predicted and equal η for the recorded holograms could not be achieved. The decreased η may be a result of partially erasing previously recorded holograms. The reduced diffraction efficiency of the holograms recorded at later times may also be caused by a gradual consumption of photoactive PQ molecules during irradiation.

Therefore, if uniform exposure doses (H) are applied to record all the holograms (in our case equal to the optimal value of H for a single hologram recording [11]), successive holograms experimentally showed a decline in diffraction efficiency (Fig. 1(d–f)).

To reduce the spatial distortion effect of further recordings on previously recorded holograms, gratings should be individually enhanced by heating of the sample after each recording step. Following this procedure we recorded a set of multiplexed reflection holograms and compared their selectivity curves (see Fig. 2).

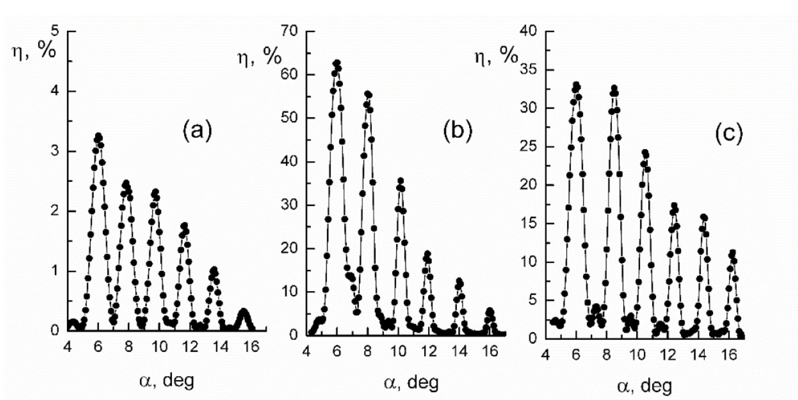

Fig. 2. Experimental curves of angular selectivity for 6 angular-multiplexed holograms (a) without and (b) with additional heating stage between recording and (c) recorded with a schedule deduced.

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This annealing process is seen to increase the diffraction efficiency value significantly, providing an amplification factor of more than 10 (compare (a) and (b) in Fig. 2). However, a decline of the diffraction efficiency at successive recordings remained. As discussed above, such non-uniformity in the values of η can be attributed to equal exposure doses upon recording. In the case of restricted number of photoactive units (molecules of PQ) optimal irradiation condition for the first hologram transform most of the PQ molecules thereby reducing the efficiency of successive holograms. The obvious solution is to exposure with a variable dose to achieve comparably uniform diffraction efficiencies.

4. Exposure-scheduling procedure

Most of the exposure schedules proposed in [3,4] are based on plotting of running curve of the cumulative grating strength (*A*) with the respect to the exposure energy (*E*). The cumulative grating strength is calculated as a sum of square roots of the measured diffraction efficiencies corresponding to the total change in the refractive index of the material:

$$A = \sum_{i=1}^{N} \sqrt{\eta_i}. \tag{2}$$

In order to derive an exposure schedule that equalizes the recorded holograms the recording curve is fitted to an analytical expression of the following form [3]:

$$A(E) = A_{sat}\left[1 - exp\left(-\frac{E}{E_\tau}\right)\right], \tag{3}$$

where E_τ is the characteristic exposure energy constant and A_{sat} is the saturation value of the grating strength, that represents so-called dynamic range of holographic polymer material. Equally strong holograms can be achieved by allocating the entire dynamic range of the material equally among the holograms, yielding [3]:

$$H_n = \frac{E_\tau}{N}\left(\frac{1}{E_\tau}\sum_{i=1}^{n-1} E_i\right), \tag{4}$$

where E_i is the amount of energy that holographic material received in order to record the i^{th} hologram, H_n is the exposure time of the n^{th} hologram. The final diffraction efficiency of each hologram can be calculated as [3]:

$$\eta_{fin} = \left(\frac{A_{sat}}{N}\right)^2. \tag{5}$$

Thus as it is seen from Eq. (4), by a special exposure schedule the recording exposure dose for each hologram can be pre-determined as well as their diffraction efficiencies (Eq. (5)). Application of Eqs. (2)–(3) in order to calculate the exposure schedule results in angle-multiplexing of 6 gratings recorded in PQ-PMMA layer (demonstrated in Fig. 3). The cumulative grating strength values were obtained from the experimental data (Fig. 2(c)).

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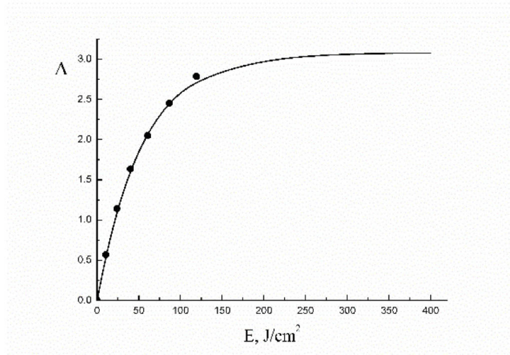

Fig. 3. The cumulative grating strength as a function of exposure energy (points) and its approximation curve (line) calculated by Eq. (2).

The parameters A_{sat} and E_{τ} of the approximation curve (Fig. 3) were 3.08 and 53.30 J/cm^2 respectively, and the predicted value of η was 26.4%. The values of H_n, calculated by Eq. (4) are given in the Table 1.

Table 1. Exposure doses for angular-multiplexed holograms' recording

N		1	2	3	4	5	6
H_n, J/cm^2	initial equal values	10.52	10.52	10.52	10.52	10.52	10.52
	calculated by Eq. (4)	8.88	10.82	13.84	18.84	27.78	45.15

The results of calculations (Table 1) show that, firstly, the exposure dose must increase with the serial number of the hologram, and, secondly, the recording of the first hologram is allocated less exposure time *H* than previously. Such intentional decreasing of H_1 seems justified to equalize the consumption of photoactive PQ molecules in the course of recording. The experimental diffraction efficiencies of 6 gratings recorded in accordance with the schedule are shown in Fig. 2(c). This implies that it is suitable to use the compensation exposure schedule for multiplexed holograms to equalize their diffraction efficiencies, though some inaccuracies remained.

5. Testing of multiplexed holograms recorded in PQ-PMMA for microscopic applications

An optimization procedure for the recording of multiplexed holographic elements in PQ-PMMA as described above was applied to VHIS with the aim to reconstruct images from different depths of the sample. For simplicity we tried to record two superimposed holograms, changing both the divergence of signal beam and the incident angle of the reference one (see Fig. 4(a)). The angle between reference and signal beams was 40 deg., NA = 0.6. To realize experimentally such type of multiplexing an objective was used in the setup and the divergence of the signal beam was controlled by its replacement. Upon reconstruction, the light waves coming from two different points within the sample are diffracted at two different angles with the respect to the optical axis, and projected onto a single image plane by a tube lens (see Fig. 4(b)).

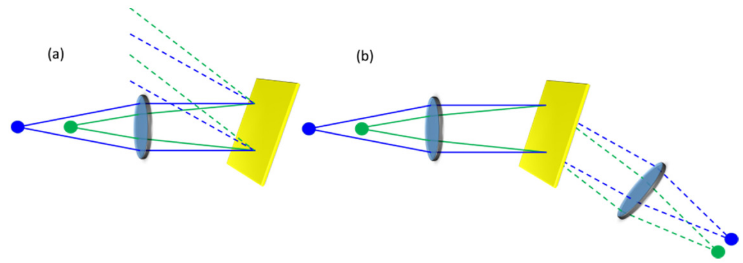

Fig. 4. Scheme (a) of the recording and (b) of the reconstruction of multiplexed holograms.

Several multiplexed holograms formed by two sequential procedures of their recording (see Fig. 4(a)) and thermal treatment were recorded in the 100 µm layer of PQ-PMMA composite. The typical dependence of the diffraction efficiency of the recorded holograms on the incident angle of the reconstructing beam is shown in Fig. 5(a). The experimental data showed that the distance between diffracted beams is about 2° being enough for their separation and independent registration.

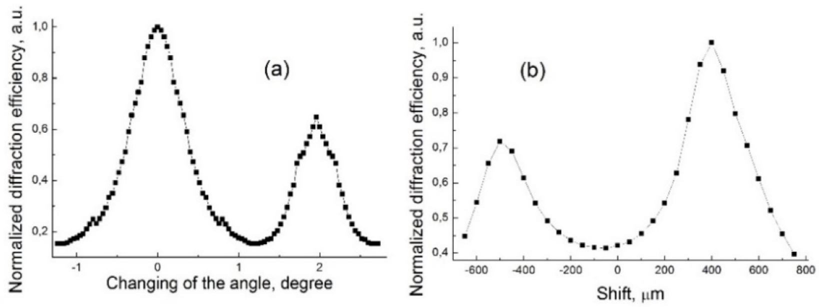

Fig. 5. Normalized (a) angular and (b) axial selectivity curves of the recorded multiplexed holograms.

The multiplexed hologram for selective excitation composed of two superimposed holograms was created by exposure of PQ-PMMA layer with signal waves of different divergence, as in scheme in Fig. 4(a), but using the same reference wave (Fig. 6(a)). Reconstruction was performed by a reference wave, resulting in two diffracted beams with different divergence (Fig. 6(b)). It is possible to create several caustics (Fig. 7) by focusing a beam by the lens or objective and to realize a selective sample excitation. The beam with lower divergence will focus first (Fig. 7(a)), after which it will start to diverge, while the second beam converges (Fig. 7(b)). At some point there will be a position when their sizes will be equal (Fig. 7(c)). Finally, the second beam focuses (see Fig. 7(e)).

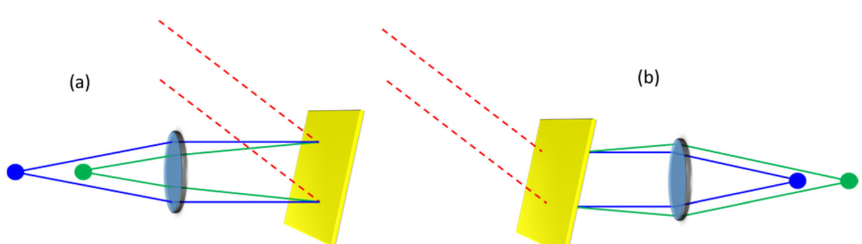

Fig. 6. Scheme (a) of recording and (b) of reconstruction of holograms for selective excitation.

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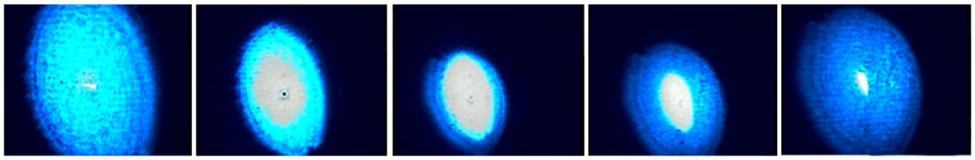

a *b* *c* *d* *e*

Fig. 7. Photos of the light beams, diffracted by the recorded hologram for selective excitation at different distances from focal plane of the focusing lens with focus f equal to: (a) −0.4f; (b) −0,2f; (c) 0f; (d) 0,2f; (e) 0,4f (f is the focal length).

To assess the reproducibility of the recorded holograms the reconstruction of them by a signal light beam with different divergences (by changing the position of objective) was performed. As it is appeared (Fig. 5(b)), the distance between curves of 1 mm corresponds to the value of shifting of the objective, which was performed by controlling the beam divergence upon holograms recording.

6. Summary

Thus, our theoretical and experimental studies of angular multiplexed holograms in the layers of polymethylmethacrylate with a high concentration of phenanthrenequinone allowed us to optimize the recording conditions. A method of equalizing the diffraction efficiencies of the recorded holograms was implemented by varying the exposure dose at each sequential recording. Multiplex hologram with the values of the diffraction efficiency for each hologram in the range of 10 to 30% were successfully developed. The possibility of practical application of the recorded multiplex holograms for the tasks of 3D-microscopy, for example for selective excitation of spatially separated regions of microscopic objects, was demonstrated in a proof of concept experiment.

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