

Experimental study of high-order reflections on volume reflection holographic gratings

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Abstract. Samples of volume reflection gratings with a wide range of periods are obtained using a holographic setup with prism coupling of radiation into the recording layer of a photopolymer material. High-order reflections on these gratings are experimentally detected. The dependence of the characteristics of observed reflections on the grating parameters and recording conditions are discussed, and the experimental and calculated wavelengths of the reflections are compared.

Keywords: volume holograms, Bragg diffraction, reflection gratings, photopolymers.

1. Introduction

Physical and chemical mechanisms of holographic grating formation in photopolymer materials (PPMs) have been studied for long time and from different points of view [1–4]. It is accepted that the key role is played by the processes of monomer diffusion and photopolymerisation. However, many aspects of the issue important for practice remain scarcely studied. In particular, the authors of Refs [5–7] discussed the conditions of the appearance of higher orders of Bragg diffraction from volume transmission holographic gratings. The phenomenon is explained by the fact that in these gratings at the final stage of their formation the profile of the periodic refractive index distribution essentially differs from a sinusoidal one, inherent in the photopolymer illumination pattern. This is due to the nonlinear dependence of the produced polymer mass on the illumination intensity, the great difference between the rates of monomer diffusion and photopolymerisation, as well as the saturation of the refractive index change with the growth of the illumination energy.

The experimental studies in this field, on the one hand, check the theoretical assumptions and, on the other hand, provide the data specifying the details of occurring processes. Based on the measurements of diffraction efficiency (DE) of such gratings in a wide range of reconstruction angles, the authors of Refs [5–8] found that the grating DE of high orders increases if the rate of monomer diffusion is smaller than that of polymerisation, as well as with increasing illumi-

nation intensity and for gratings with the period smaller than 1500 lines mm^{-1} . In Refs [5, 6] the estimates of the refractive index modulation for the fundamental grating and its second harmonic were obtained, their ratio found to be 4:1 and higher.

In Ref. [8] devoted mainly to the use of the second diffraction order from volume transmission gratings in Bayfol HX photopolymers (Bayer Material Science AG, Germany) for producing planar waveguides, the model and experimental evidence was found that when the illumination intensity and energy exceed 10 mW cm^{-2} and 100 mJ cm^{-2} , respectively, the DE considerably decreases for the first diffraction order and, on the contrary, essentially grows for the high diffraction orders.

Note that the authors of Ref. [8] modelled the refractive index distribution profile in reflection gratings having a period of ~ 4500 lines mm^{-1} in these materials and demonstrated the absence of deviation from a sinusoidal pattern. However, this conclusion is not confirmed experimentally.

At the same time, there are publications in which the existence of high orders of diffraction from the volume reflection holographic gratings is not only assumed theoretically [9], but also experimentally observed [10] and explained by the influence of the refractive index remodulation in BB640 silver halide emulsions. Unfortunately, the authors of paper [10] could not demonstrate the presence of these harmonics in the single transmission spectrum of the studied gratings. Obviously, it is related to the complexity of such experiments, since it is necessary to record the reflections, significantly separated from each other in the spectral range 250–1500 nm with high resolution. In Ref. [10], this problem was avoided by artificially increasing the grating period using the technology of gelatine swelling in water.

The purpose of the present paper is the experimental study of high-order reflections on volume reflection holographic gratings in PPMs, including the grating fabrication and the measurement of diffraction and spectral characteristics. These data can be useful both for specification of hologram formation mechanisms in photopolymer materials and for their application in spectral filters, displays, security holograms, etc.

2. Theoretical aspects

High orders of Bragg diffraction from volume holographic gratings arise because the profile of the resulting refractive index distribution in these gratings essentially differs from the sinusoidal one and has the form [5]:

$$n(x) = n_0 + \sum_{i=1}^{\infty} n_i \cos\left(\frac{2i\pi x}{\Lambda}\right), \quad (1)$$

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where x is the spatial coordinate along the grating vector; $i = 1, 2, 3, \dots$; $\Lambda = \lambda_0 / (2n_0 \cos \theta_1)$ is the fundamental grating period; λ_0 is the laser radiation wavelength (in air) used to record the grating; n_0 is the mean refractive index of the recording medium; and θ_1 is the Bragg angle in this medium. In particular, n_1 , n_2 and n_3 are the amplitudes of the photopolymer refractive index modulation, corresponding to the fundamental grating, its second and third harmonics.

If a beam of white light is incident on the volume grating normally to its surface, then, according to the theory of Ref. [11], the dips with centre wavelengths $\lambda_i^r = 2n_0 \Lambda_i = 2n_0 \Lambda / i$ must appear in the spectrum of the transmitted light due to the Bragg reflection.

Using the line of reasoning analogous to [5], where the expressions for the DE of the volume transmission grating and its second harmonic based on the coupled wave theory [11] are presented, we can suppose that for the volume reflection grating and its harmonics the following formula is valid

$$\eta_i^r = \tanh^2 \frac{\pi n_i T}{2n_0 \Lambda_i}, \quad (2)$$

where η_i^r is the DE of the corresponding grating.

From the known experimental values of η_i^r one can find n_i and their ratios, in particular

$$n_1 = \frac{2\Lambda n_0 \tanh^{-1}(\sqrt{\eta_1^r})}{\pi T},$$

$$\frac{n_1}{n_2} = 2 \frac{\tanh^{-1}(\sqrt{\eta_1^r})}{\tanh^{-1}(\sqrt{\eta_2^r})}, \quad \frac{n_1}{n_3} = 3 \frac{\tanh^{-1}(\sqrt{\eta_1^r})}{\tanh^{-1}(\sqrt{\eta_3^r})}. \quad (3)$$

In studying the spectral characteristics of the fundamental grating and its harmonics, one should keep in mind that the mean value of the refractive index of the recording medium n_0 increases with increasing illumination energy and then saturates. An increase in n_0 is accompanied by a decrease in the PPM volume (mainly the layer thickness) and is known as shrinkage [12]. The influence of the PPM refractive index dispersion is also essential, due to which the values of n_0 for the reflections in the long-wavelength and short-wavelength regions of the spectrum are different. Specific features of these processes in application to the subject of study will be considered below.

3. Experimental studies

3.1. Setup for recording the volume reflection gratings

Volume reflection gratings are usually formed using the interference of two crossing laser beams. The grating period depends on the wavelength of the used radiation, the refractive index of the recording medium, the convergence angle of the beams (usually from 60° to 180° in air), and can amount to 0.16 – $0.26 \mu\text{m}$ for the photopolymer materials with $n_0 \sim 1.5$ and the laser radiation with the wavelengths from 473 to 633 nm . In Ref. [13] the volume reflection gratings with the period $0.24 \mu\text{m}$ were recorded using the radiation at the wavelength 514 nm and the recording scheme with two rectangular optical prisms coupling the radiation into PPM and providing the convergence angle 90° of recording beams (in the medium). We developed a setup with some elements adopted from Ref. [13], but the scheme is significantly simplified and comprises only one coupling prism.

The optical scheme of our setup is presented in Fig. 1 (the transverse section is shown). By means of the optical elements not presented in the figure, the s -polarised collimated light beam is formed, below referred to as a reference beam. It is normally incident onto the narrow lateral face of the optical prism made of BK7 glass, its base having the shape of an isosceles right-angled triangle. This beam sequentially propagates through the prism and the PPM film with adhesive properties, glued by one surface to the wide lateral face of the prism and by another surface to the upper surface of the transparent substrate (made of plastic or glass). Since the angle of incidence on the substrate amounts to 45° and exceeds the critical angle of total internal reflection, equal to 41.3° for the substrate material with the refractive index ~ 1.5 , the beam experiences total reflection from the bottom surface of the substrate contiguous with air. The reflected beam (in fact, an object one) preserves the s -polarisation and interferes with the reference beam, providing the recording of a volume reflection holographic grating in the PPM.

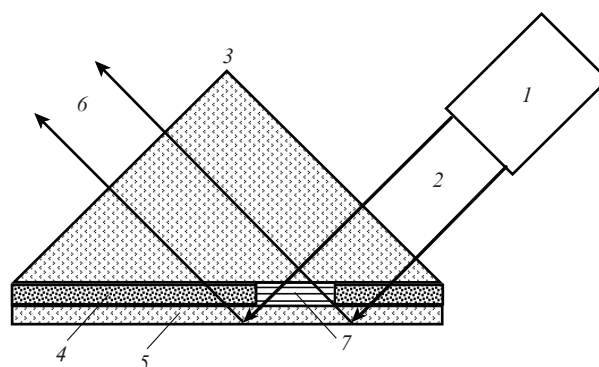


Figure 1. Schematic diagram of the setup for recording volume reflection gratings: (1) laser; (2) collimated light beam; (3) optical prism; (4) photopolymer material; (5) optical substrate; (6) reflected (object) beam, necessary for the formation of the volume reflection holographic grating (7).

In order to extend the range of periods of the studied gratings, we also used a dispersion prism made of glass with the refractive index 1.515 and having the apex angle 67° . The angle of the reference beam incidence on the lateral face of this prism was chosen such that in the PPM medium it would be close to 65° . In this case, the calculated wavelengths of Bragg reflections of the fundamental grating, its second and third harmonic amount to ~ 1500 , 750 , and 500 nm , respectively, which is interesting for constructing optical communication devices, spectral filters, and displays.

The measurement of spectral characteristics of the studied gratings was carried out using a SHIMADZU UV/VIS 2501 PC spectrometer with a double monochromator (Shimadzu Corporation, Japan) and a Yokogawa AQ6370 optical spectral analyser (Yokogawa, Japan), operating in the ranges 190 – 1100 nm and 600 – 1700 nm , respectively, with the resolution 0.1 nm .

Using the spectroscopic method [14], the DE values of the volume reflection gratings were estimated as

$$\eta^r = (1 - Tr_r / Tr_0) \times 100\%, \quad (4)$$

where Tr_r is the transmission coefficient of the hologram near the minimum of the spectral dip, caused by the reflection of

light; Tr_0 is the transmission coefficient beyond this region (near the base of the contour).

3.2. Discussion of experimental results

Several samples of volume reflection gratings were recorded. Gratings 1a and 1b were recorded using a He–Ne laser (the radiation wavelength 632.8 nm), grating 2 using a DPSS laser (532 nm) (in all these cases the optical prism with the isosceles right-angled triangle base was used), and grating 3 using the He–Ne laser and the spectroscopic prism mentioned above. The choice of the grating parameters was determined by the purpose to detect the reflections of the fundamental grating and its harmonics in the wide region (300–1500 nm) of the spectrum. In the experiments, we used a BAYFOL HX TP colour photopolymer belonging to the line of Bayer MaterialScience AG products [4].

In the case of the He–Ne laser used for gratings 1a and 3, the intensity of the reference beam was 15 mW cm^{-2} and the recording energy was 250 mJ cm^{-2} . For grating 1b, the intensity was 3 mW cm^{-2} and the recording energy was the same. For the DPSS laser (grating 2) the intensity was 6 mW cm^{-2} and the energy was 150 mJ cm^{-2} . The power of laser radiation was measured using a VEGA device with photoelectric sensor (OPHIR, Israel). After recording, the samples of PPMs with gratings were detached from the prisms and illuminated with UV radiation, according to [4], in order to bleach the residual photopolymer dye. Then a beam of probing white light was directed on the gratings normally to their surfaces, and the transmitted light was recorded with the above spectral instruments.

Note that the parasitic multiple reflections were eliminated by choosing the angle of incidence of the reference beam on the prism with the right-angle triangle base to be actually equal to $\sim 46^\circ$. With the refractive index of the prism glass (~ 1.515) and the photopolymer (~ 1.5) taken into account, the angle of propagation of the reference beam in the PPM could be estimated as nearly 46.3° . Below this circumstance is taken into account in the calculation of the wavelength for the reflections of the fundamental grating and its harmonics.

Figure 2 presents the transmission spectrum of the volume reflection grating 1a, fabricated using the He–Ne laser and the above setup. The narrow dips caused by the Bragg reflection of light are well seen. Alongside with the reflection corresponding to the fundamental grating, the reflections belonging to its second and third harmonic are present. As a result of the experimental data processing, it is found that the fundamental grating reflection wavelength is $\lambda_1^r \sim 904 \text{ nm}$, the contour width is $\Delta\lambda_1^r \sim 8 \text{ nm}$ (at the level of 50% of the depth). The DE η_1^r calculated using the formula (4) amounts to $\sim 50\%$. For the second harmonic $\lambda_2^r \sim 457 \text{ nm}$, $\Delta\lambda_2^r \sim 4 \text{ nm}$, and $\eta_2^r \sim 17\%$. For the third harmonic $\lambda_3^r \sim 313 \text{ nm}$, $\Delta\lambda_3^r \sim 1 \text{ nm}$, and $\eta_3^r \sim 3\%$. Note that the measurement errors did not exceed $\pm 0.1 \text{ nm}$ for the wavelengths of reflections and $\pm 1\%$ for the diffraction efficiency. According to formula (3), it was found that $n_1 \sim 0.005$, $n_1/n_2 \sim 4$, $n_1/n_3 \sim 15$, i.e., the fraction in the spectrum of reflected light is essential for the second harmonic and small for the third one.

Figure 3 presents the transmission spectrum of the volume reflection grating 1b. In contrast to Fig. 2, here $\lambda_1^r \sim 902 \text{ nm}$, the DE of the fundamental grating η_1^r is greater and amounts to $\sim 66\%$; for the second harmonic $\lambda_2^r \sim 456 \text{ nm}$, $\eta_2^r \sim 7\%$; for

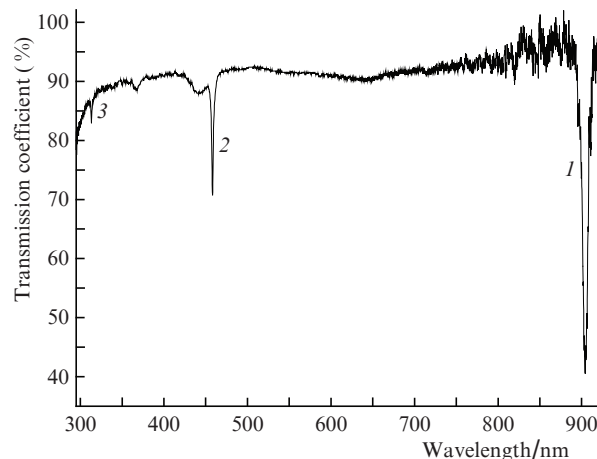


Figure 2. Transmission spectrum of the volume reflection grating 1a: (1) the fundamental grating reflection; (2) and (3) the second and third harmonic reflections.

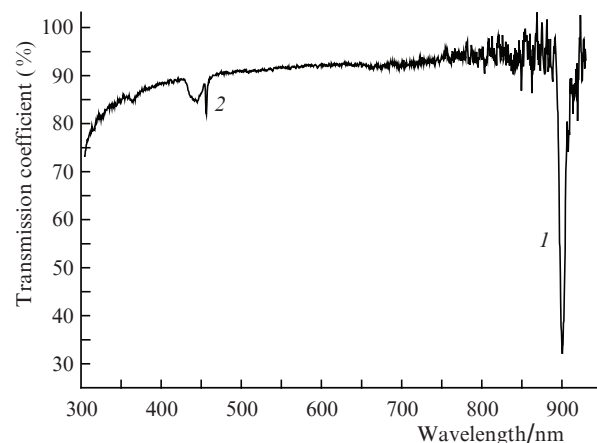


Figure 3. Transmission spectrum of the volume reflection grating 1b: (1) the fundamental grating reflection; (2) the second harmonic reflection.

the third harmonic the reflection is absent or not recognised against the noise background.

Thus, the theoretical assumption [8] that the high-order DE falls with decreasing illumination intensity (provided that its energy is conserved) is experimentally confirmed.

The transmission spectrum of grating 2, fabricated at the same setup but with the DPSS laser (532 nm) is shown in Fig. 4. It is seen that for the fundamental grating the reflection wavelength is $\lambda_1^r \sim 754 \text{ nm}$, the contour width (at 50%-depth level) is $\Delta\lambda_1^r \sim 6 \text{ nm}$, the DE calculated using formula (4) is $\eta_1^r \sim 70\%$. For the second harmonic $\lambda_2^r \sim 384 \text{ nm}$, $\Delta\lambda_2^r \sim 2 \text{ nm}$, and $\eta_2^r \sim 6\%$. In this case, the fraction of the second harmonic in the spectrum of the reflected light is insignificant. The reflection of the third harmonic with the expected wavelength $\lambda_3^r \sim 255 \text{ nm}$ (i.e., in the UV region where the absorption in the PPM is more significant) could not be detected.

Figure 5 presents the transmission spectrum of the volume reflection grating 3, fabricated using the He–Ne laser and the spectroscopic prism mentioned above. It is a result of matching two spectra obtained from the same grating by means of different spectral instruments. The experimental points of the spectrum recorded using the Yokogawa AQ6370 spectrum

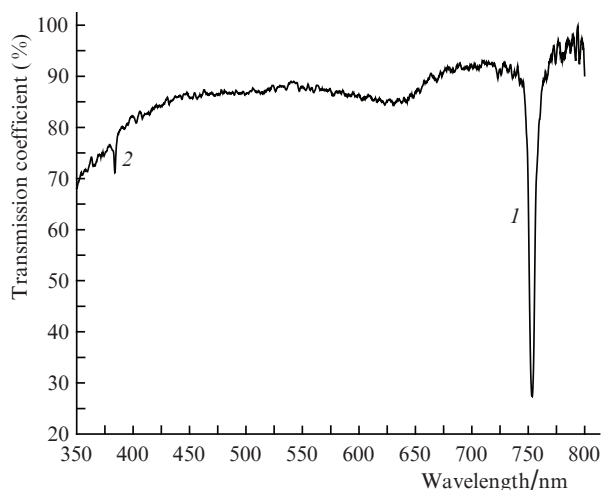


Figure 4. Transmission spectrum of the volume reflection grating 2: (1) the fundamental grating reflection; (2) the second harmonic reflection.

analyser are shown by the light circles and the experimental points obtained with the SHIMADZU UV/VIS 2501 PC spectrophotometer are shown by the dark ones. The matching was possible because the reflections of the second harmonic of the studied grating coincided in both spectra, which is well seen in the magnified fragment.

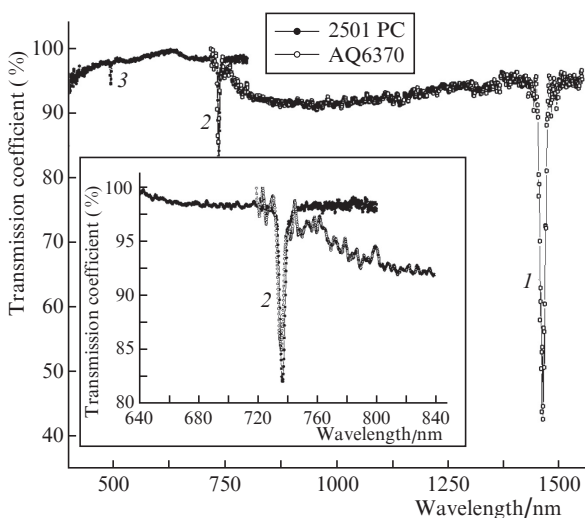


Figure 5. Transmission spectrum of the volume reflection grating 3: (1) the fundamental grating reflection; (2) and (3) the second and third harmonic reflections.

After the data processing, it was found that the fundamental grating reflection wavelength is $\lambda_1^r \sim 1462$ nm, the contour width (at the level of 50% of the depth) is $\Delta\lambda_1^r \sim 12$ nm, the DE calculated using formula (4) is $\eta_1^r \sim 53\%$. For the second harmonic $\lambda_2^r \sim 736$ nm, $\Delta\lambda_2^r \sim 4$ nm, and $\eta_2^r \sim 15\%$; for the third harmonic $\lambda_3^r \sim 495$ nm, $\Delta\lambda_3^r \sim 1$ nm, and $\eta_3^r \sim 3\%$. Then, according to formula (3), $n_1/n_2 \sim 4.5$, $n_1/n_3 \sim 16$, i.e., the fraction of the second harmonic in the spectrum is considerable and the fraction of the third harmonic is insignificant.

Note that the experimental wavelengths of reflections λ^r for all studied gratings differ from the calculated (theoretical) ones $\tilde{\lambda}^r$, which can be found using the formulae of Ref. [15].

These differences are caused by a change in Bragg diffraction conditions, relating the period of the volume reflection grating Λ , the refractive index n , and the reflection wavelength as [11]

$$\tilde{\lambda}^r = \Lambda 2n. \quad (5)$$

Such changes arise in the process of grating fabrication and PPM post-processing due to the so-called effective shrinkage of the photopolymer S_{eff} [15, 16], allowing for both the reduction of its thickness T (geometric shrinkage) and the increase in the mean value of n . The geometric shrinkage $\Delta T/T$ achieving 0.01–0.02 [15, 17] leads to the proportional reduction of the grating period and, therefore, to the shift of the reflection towards the short-wavelength region of the spectrum. On the contrary, the increase in the photopolymer refractive index $\Delta n/n$ amounting to 0.005–0.01 [7, 17] shifts the reflection wavelength towards the long-wavelength region.

Note that the ratios $\lambda^r/\tilde{\lambda}^r$ for the reflections of the fundamental grating and its harmonics are expected to be similar, since the expression

$$\frac{\lambda^r}{\tilde{\lambda}^r} = S_{\text{eff}} = \left(1 + \frac{\Delta n}{n}\right) \left(1 - \frac{\Delta T}{T}\right). \quad (6)$$

is valid, independent of the grating period.

However, the calculations show that for the wavelength $\lambda = 633$ nm and the PPM refractive index $n = 1.5$ the period of the fundamental grating 1a equals 305.4 nm, the wavelength of its reflection is $\tilde{\lambda}_1^r = 916$ nm, and for the second and third harmonic it is 458 and 305 nm. In this case, the ratios of experimental and calculated values of the reflections appear to be different, namely 0.9869, 0.9978, and 1.0262, respectively.

To remove this inconsistency, let us notice that according to Eqn (5) the calculation of the wavelength of a grating reflection should be performed with the dispersion of the PPM refractive index $n(\lambda)$, i.e., the dependence on the observation wavelength taken into account. The necessary data for the Bayer MaterialScience photopolymers are presented in Ref. [17], from which it follows (with extrapolation) that in the interval of wavelengths 900–300 nm the mean value of the refractive index varies from 1.490 to 1.545, which considerably differs from the constant value $n = 1.5$ used by us above. The appropriate correction yields the new calculated values for the grating 1a $\tilde{\lambda}_1^r = 910$ nm, $\tilde{\lambda}_2^r = 460$ nm, and $\tilde{\lambda}_3^r = 315$ nm; the ratios of these values and the experimental ones are 0.9934, 0.9935, and 0.9936.

Thus, we found an adequate explanation for the difference between the experimental and calculated values of the wavelengths of reflections for grating 1a. It consists in the fact that both the effective shrinkage of the PPM and the dispersion of its refractive index are to be taken into account. Analogous calculations can be performed for the gratings considered above.

4. Conclusions

Using the setup with prism coupling of radiation into the recording layer of photopolymer material, we obtained the

samples of volume reflection gratings within the range of periods 2000–4000 lines mm^{-1} .

The reflections of the fundamental gratings, their second and third harmonics were experimentally recorded. In particular, for the grating with the fundamental reflection at the wavelength 904 nm the harmonics at the wavelengths 457 and 313 nm were observed, their DE being $\eta_1^r \sim 50\%$, $\eta_2^r \sim 17\%$, and $\eta_3^r \sim 3\%$, respectively.

For the photopolymers in question, the presence of high-order reflections weakly depends on the period of reflection gratings. The main role is apparently played by the ratio of the monomer diffusion rate and photopolymerisation one, as well as the saturation of the photoinduced change of the PPM refractive index at large values of the illumination intensity and energy. The theoretical assumption that for high-order reflections the DE grows with the increase of the illumination intensity is confirmed. At the same time, it is clear that the issue requires deeper investigation, correlated with the particular composition of the PPM.

The comparison of experimental and calculated values of the wavelengths of reflections should be carried out considering both the effective shrinkage of the PPM and the dispersion of its refractive index.

The obtained results can be used to specify the mechanisms of hologram formation in photopolymer materials, as well as for practical applications in spectral instrumentation, holographic optical elements, displays, etc.

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