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Polarization Holographic Gratings Formed on Polymer Dispersed Liquid Crystals

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1. Introduction

Development and investigation of new active optical media purposed for the reflection of optical information and also for the control of light propagation direction and properties in photonic devices remain essential under the conditions of rapid development of information technologies. Because of their optically active properties, polarization holographic gratings are promising in the production of high-function optical devices such as lights modulators, valves, polarization multiplexers and demultiplexers, polarized radiation splitters (Bunning et al., 2000; Cipparrone et al., 2000; Cipparrone et al., 2001a; Habraken et al., 1995; Ono et al., 2005; Wu et al., 2001; Yu et al., 2002). Such gratings are recorded on polarization-sensitive materials: photo-refractive crystals (Sturman et al., 1996), «guest-host» systems (Khoo, 1991; Slussarenko et al., 1997; Nikolova & Todorov, 1984), azopolymers (Naydenova et al., 1998; Labarthet & Rochon, 1999). The reports about the record of the polarization gratings in liquid-crystal composites (LC-composites) which do not contain azodyers, are comparatively recent (Cipparrone et al., 2001a; Mazzulla et al., 2004).

The LC-composites present a polymer matrix, within which the droplets of nematic liquid crystals (NLC) are dispersed. They are appealing by the fact that they combine the positive features of polymers (mechanical strength, stability, flexibility, low price) and unique physical peculiarities of liquid crystals (anisotropy of optical and dielectric properties). Such composite materials allow using a relatively simple one-stage technology of the optical record, a long-term retention of the polarization state, which provides the high polarization selectivity of the recorded polarization gratings. Moreover, the diffraction efficiency of the polarization holographic gratings recorded in the LC-composites can be controlled by electric and thermal fields.

The polarization holographic gratings form in a thin LC-composite layer as a result of the NLC-polymer phases separation during the photopolymerization caused by the superposition in the plane of the sample of two coherent waves polarized in the mutually perpendicular planes or two left- and right-circularly polarized waves. In this case, the light modulation is absent, and the changing phase difference between interfering waves results solely in the variation of the polarization state of the total light field. Since the NLC is sensitive to the total field direction and is able to orient in the light-field vector direction, a

grating with repeating orientations of the NLC molecules in polymer droplets forms in the LC-composite after the exhibition.

The functionality of the formed gratings, effective control of their electric-optical properties depend on the NLC physical properties and are also related with the peculiarities of the supramolecular structure which is realized in the forming composite. Among the parameters governing the supramolecular structure, the following should be noted: the NLC and polymer type, LC – polymer ratio, technological process peculiarities (record energy, formation temperature).

Because of big amount of components in the pre-polymer system, it is necessary to study the balance between their concentrations and corresponding physical and chemical properties of the forming gratings, which dictate the grating controllability by the electric field. The influence of all mixture components on the properties of reflective and transmission gratings formed at the light photopolymerization with the modulated intensity has been described in the literature (Beev et al., 2006; Zharkova et al., 2007). In these cases, the periodical supramolecular structures form at competing processes of photopolymerization, diffusion, phase separation of the NLC and polymers and vitrification. When the polarization gratings form, the initial composition is illuminated by the light of constant intensity thus there are no concentration gradients of the components in dark and light bands of the interferential pattern. Therefore the processes of substance diffusion are absent. That is why the quality of the polarization gratings depends on the photopolymerization, NLC - polymer phase separation and vitrification. These processes significantly depend on the initial composition content and record conditions. Today, for the polarization gratings, there are no clear conditions of optimization of all processes involved in the formation of gratings with the maximum diffraction efficiency because of the lack of experimental data. The reason is that for each individual system there are its optimum component relations and exhibition conditions.

The peculiar structure of the NLC in such materials presumes the dependence of the polarization holographic gratings efficiency on the incident radiation polarization, as well as the difference in the polarization states of the diffracted and non-diffracted radiation, which enables to control the direction of the optical radiation propagation and polarization.

Since the anisotropic properties of the nematic liquid crystal, which is the component of the pre-polymer composition, are influenced by the temperature and external electric field, the peculiarities of thermal-optical and electric-optical switching of the polarization holographic gratings are of interest.

Thus, the purpose of this work is to study the influence of the formation conditions of the polarization holographic gratings, recorded in the LC-composites, on their diffraction efficiency to investigate the polarization selectivity of such gratings, as well as to study the peculiarities of the thermal-optical and electric-optical switching of the polarization holographic gratings and to demonstrate the possibility of using them as light modulators.

2. Theory

The polarization gratings record resulted from the superposition of two plane waves with mutually orthogonal polarizations and equal amplitudes E_0 . The waves were polarized as follows: a) at the angles of +45^o and -45^o to the incidence plane; b) at the angles of 0^o and 90^o to the incidence plane (*s*- and *p*-polarization) (Fig.1).

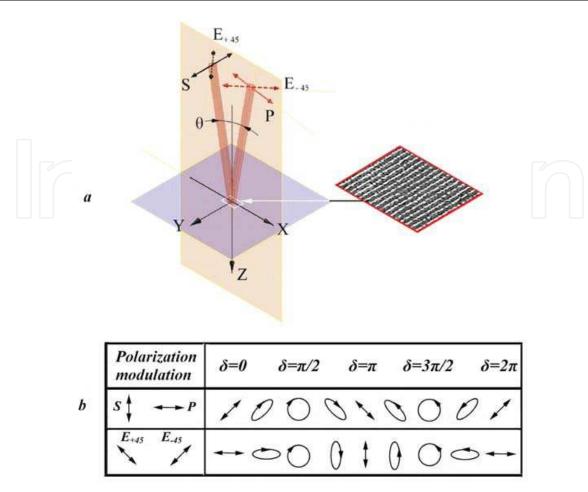


Fig. 1. Geometry of polarization holographic gratings record (a) and respective spatial polarization modulation along the axis *x* (b). The photo of the polarization holographic grating is obtained by the polarization microscopy method, polarizers are crossed.

The total field strength $E=E_1+E_2$ can be written with the Jones vector, for the case a):

$$E = \frac{2E_0}{\sqrt{2}} \cdot \begin{bmatrix} \cos\frac{\theta}{2} \cdot \cos\frac{\delta}{2} \\ i \cdot \sin\frac{\delta}{2} \end{bmatrix};$$
(1)
where
$$\delta = \frac{4\pi \cdot x \cdot \sin(\frac{\theta}{2})}{\lambda};$$
(2)

W

is the difference of interfering waves phases; λ is the wave length; θ is the angle of interfering waves convergence;

$$E = E_0 \cdot \begin{bmatrix} \cos\frac{\delta}{2} \\ i\sin\frac{\delta}{2} \end{bmatrix};$$
 (3)

for the case b):

Let us presume that at the superposition of two waves with orthogonal polarizations, the varying refractive index gives the major contribution into the polarization grating formation. Moreover, presume that only the linear anisotropy of the refractive index takes place for the used composition at the grating record.

In this case, the Jones matrix, which describes the transmission and diffraction from the recorded grating, can be determined as follows (Nikolova & Todorov, 1984):

where
$$T_{\mathbf{1}} = \begin{bmatrix} exp\{(i\Delta\phi)cos\delta\} & 0\\ 0 & exp\{-(i\Delta\phi)cos\delta\} \end{bmatrix};$$
(4)
$$\Delta\phi = \frac{\pi\Delta nd}{\lambda};$$
(5)

 Δn is the photo-induced modulation of the refractive index; *d* is the sample thickness; While the polarization grating is forming, a surface relief may occur during the phase separation process. The incident radiation diffracts on it, too. The Jones matrix, which describes the diffraction of such a surface relief, can be written as follows (Naydenova et al., 1998):

$$T_{2} = \begin{bmatrix} exp\{(i\Delta\psi)cos(\delta + \delta_{1})\} & 0\\ 0 & exp\{(i\Delta\psi)cos(\delta + \delta_{1})\} \end{bmatrix};$$
(6)

$$\Delta \psi = \frac{2\pi \cdot d_1}{\lambda} \cdot \frac{(n_p - n_a)}{2}; \tag{7}$$

is the value of the phase modulation caused by the surface relief; d_1 is the surface modulation depth; n_p is the average refractive index of the LC-composite; n_a is air refractive index; δ_1 is the phase shift between the relief and polarization gratings.

The field of the diffracted wave can be defined as follows (Cipparrone et al., 2001b):

$$E_{out} = T_2 T_1 E_{in}; \tag{8}$$

(9)

where $E_{in} = E_{0i} \begin{bmatrix} \cos \alpha \\ \sin \alpha \end{bmatrix};$

is the Jones vector describing the incident wave; E_{0i} is the amplitude of the incident wave; *a* is the angle between the direction of the test beam polarization and wave vector of the grating. The polarization properties of the beam diffracted into the *m*-order are determined by the Fourier components $E_{diffn}(a)$ on the paraxial approximation (Tervo & Turunen, 2000):

$$E_{diffm}(a) = \frac{1}{\Lambda} \times \int_{0}^{\Lambda} E_{out}(x) exp(-i2\pi mx / \Lambda) dx;$$
(10)

where

$$\Lambda = \frac{\lambda}{2\sin(\theta/2)} \tag{11}$$

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is the grating period;

where

the diffraction efficiency of the polarization grating can be defined as:

$$\eta_m = exp(-\sigma \times d) \times \frac{\left| E_{diffm}(a) \right|^2}{\left| E_{in} \right|^2};$$
(12)

where σ is the extinction ratio.

3. Formation of the polarization gratings and experimental methods

Polarization gratings were recorded as a result of superposition of two plane waves of similar intensity with mutually orthogonal linear polarizations. In one case, the waves were polarized vertically and horizontally (s and p), in the other case – at the angle of $\pm 45^{\circ}$ (E₊₄₅ and E₋₄₅) to the incident plane (Fig. 1). At the superposition of the waves with mutually orthogonal polarizations, solely the polarization state varies in the sample plane (XZ), and the modulation intensity is absent.

Fig. 2 shows the schematic of the polarization holographic grating record. A semi-conductor laser with λ = 658 nm and power of 80 mW was used as a radiation source. The radiation power density in the sample plane was 42 mW/cm². The angle of convergence of the interfering light beams was θ = 1.3^o - 2^o. The period of the obtained gratings was equal to 19 - 30 µm, which corresponded to the spatial frequency of 53 - 34 mm⁻¹.

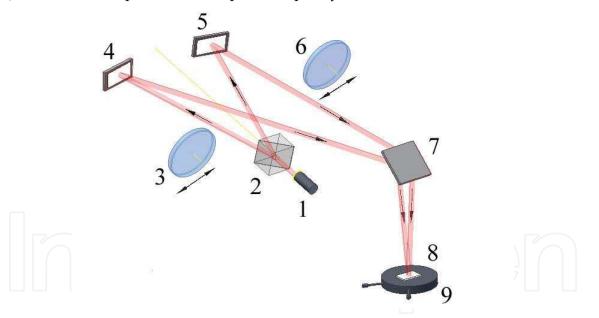


Fig. 2. Schematic of the facility for polarization holographic gratings record. 1 – laser diode ML101GR (λ = 658 nm); 2 – Wollaston prism; 3, 6 – plates $\lambda/2$; 4, 5, 7 – mirrors; 8 – sample; 9 – thermostatic table.

The initial prepolymer composition presented a mixture of an acrylate monomer, NLC, cross-linking component and the photo-initiating system which included a dye with the maximum absorption in the visible spectrum, and a co-initiator. Pentaerythritol tetraacrylate (Sigma-Aldrich) (n_p =1,487) was used as the monomer, and the mixture BL087 (Merck) (n_o =1.527, n_e =1.797, $\Delta\epsilon$ >0, $T_{\rm NI}$ =100 °C) was applied as the NLC. In the composition, N-vinylpyrrolidone served as the cross-linking component and a solvent for the dye and co-

initiator. Methylene-blue was used as the dye; it guaranteed the maximum light absorption within the wave length range of 650-680 nm. The NLC concentration in the prepolymer composition was 25-55 mass %; the dye concentration was 0.3 mass %. The composition was prepared as a homogeneous solution which was deposited between two glass slides coated with a transparent conductor. Sample thickness was 10 - 50 μ m.

At the radiation diffraction on the formed polarization gratings, we observed the high-order diffraction (Fig. 3). The Klein's parameter for the such gratings is Q:

$$Q = 2\pi\lambda d / (n\Lambda^2) \le 1;$$
(13)
(Raman-Nath regime), i.e. we deal with plane holograms.

Fig. 3. Diffraction pattern observed at the diffraction of the polarization grating.

The quality of the recorded polarization holographic gratings was evaluated by their diffraction efficiency. The diffraction efficiency of such gratings for the light diffracted into the ±1 order, was defined as $\eta_{\pm 1} = I_{\pm 1}/I_0$, where $I_{\pm 1}$ is the intensity of the light diffracted into the first order, I_0 is the incident light intensity the onto the grating. Fig. 4 shows schematically the experimental facility for the study the polarization selectivity of the polarization grating. The probe beam was linearly polarized, the direction of its polarization formed the angle α with the direction of the grating wave vector **k**.

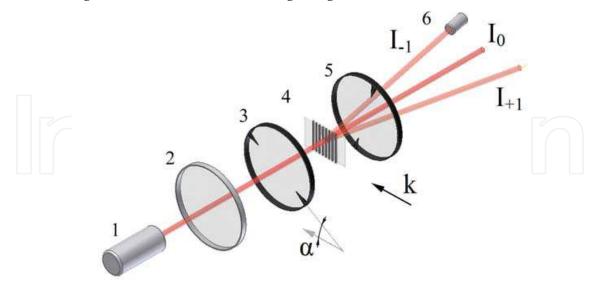


Fig. 4. Schematic of the facility to study the polarization selectivity of the polarization gratings. 1 – semiconductor laser (λ = 658 nm); 2 - $\lambda/4$ plate; 3 – polarizer; 4 – sample under study; 5 – analyzer; 6 – photodetector.

To study the thermal-optical switching, the studied sample was set into a thermostatic cell.

4. Experimental results

It is possible, varying the content of the prepolymer composition and its lighting mode, to control the kinetics of photopolymerization and components phase separation, and hence to influence the supramolecular structure of the obtained composite and, consequently, the properties of the forming polarization holographic grating. Two factors have the most significant effect on the formed composite morphology: a) the NLC – polymer ratio in the initial prepolymer composition; b) exposure energy.

The study of the effect of the NLC concentration in the initial prepolymer composition has revealed that the gratings with better efficiency form within quite a narrow NLC concentration interval (35 - 38%), and any deviation from it results in the noticeable deterioration of their quality (Fig. 5).

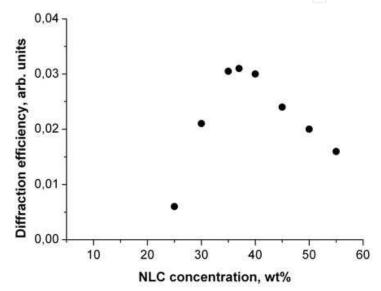


Fig. 5. Diffraction efficiency of polarization holographic gratings vs the NCL concentration in the initial prepolymer composition. Sample thickness is $20 \,\mu$ m. Spatial frequency of the grating is $34 \,\text{mm}^{-1}$.

The studied polarization gratings result from the polarization and phase separation processes. At low NLC concentrations (~ 25%), the process of the phase separation of polymer and liquid-crystal phases is at the beginning, and at the concentrations of 45 - 55% it is almost over, which causes strong light dispersion on numerous polymer-NLC interfaces and hence results in increasing dispersion noise and decreasing diffraction efficiency of the grating.

Fig. 6 presents the dependence of the diffraction efficiency of the polarization holographic grating, formed in the LC-composite on the record energy density for the grating with the spatial frequency of 34 mm⁻¹. It is evident that the diffractive efficiency rises along with the record energy up to ~3%, then the growth decelerates and saturation is observed at η = 3.5% (for the sample of 20 µm). The threshold record energy corresponding to the diffraction efficiency of 1% is 0.65 J/cm², and the total record energy at which the maximal diffraction efficiency is reached is equal to 2.5 J/cm². The sample thickness influences these values. The smaller the thickness of the LC-composite, the lower energy density is needed to finish the orientation processes of the NLC and processes of the phase separation of polymer and liquid-crystal phases at the specific radiation power density.

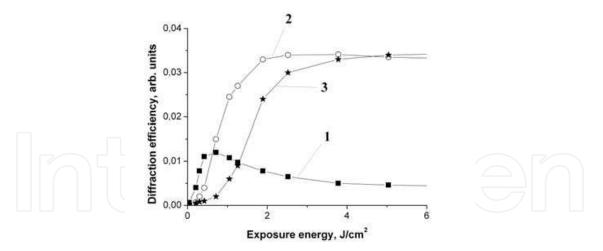


Fig. 6. Diffraction efficiency of the polarization holographic gratings vs the energy density for LC-composites of various thickness. 1 – thickness of $10 \,\mu\text{m}$; 2 – $20 \,\mu\text{m}$; 3 – $50 \,\mu\text{m}$. NLC concentration is 37 mass %. Spatial frequency of the grating is 34 mm⁻¹.

As the spatial frequency of the recorded polarization grating grows (along with the growing angle of interfering beams convergence), we observe the increase of the total energy needed for its forming, regardless the grating thickness (Fig. 7). The dependence of the diffraction effectiveness on the record energy density features the maximum, which position and value depend on the grating spatial frequency. Moreover, in our experiments, for the chosen prepolymer composition, the polarization gratings with the spatial frequency above 60 mm-1 did not form even at significant record energies.

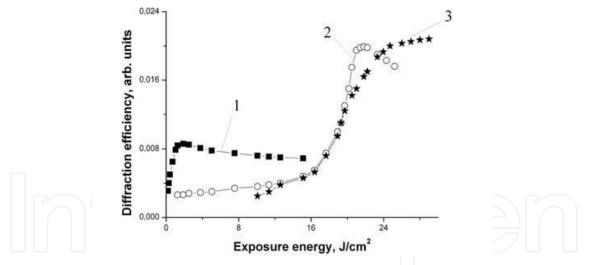


Fig. 7. Diffraction efficiency of the polarization holographic gratings vs the energy density for LC-composites of various thickness. 1 – thickness of $10 \mu m$; 2 – $20 \mu m$; 3 – $50 \mu m$. NLC concentration is 37 mass %. Spatial frequency of the grating is 53 mm⁻¹.

At the polarization grating formation, the process of phase separation is temperature-sensitive; along with the temperature growth, the NLC and monomer viscosity decrease, the phase separation of the NLC and polymer is faster and better, and the polymerization velocity increases. Moreover, the temperature influences the process of NLC orientation in the droplets. Fig. 8 shows the dependence of the diffraction efficiency of the polarization holographic gratings on the formation temperature for the temperature range: from the room temperature to the point of NLC transition in an isotropic liquid.

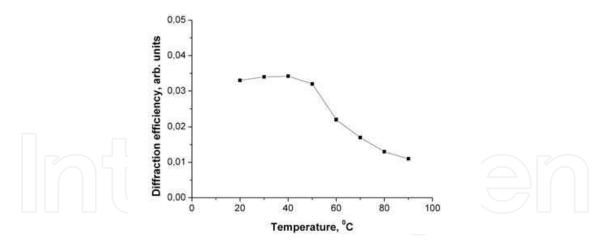


Fig. 8. Diffraction efficiency of the polarization holographic gratings vs the formation temperature. Sample thickness is 20 μm. Spatial frequency of the grating is 34 mm⁻¹.

Evident that the maximum diffraction efficiency is reached at the formation temperature of 35 - 45 °C. This is apparently the temperature range in which the processes of polymerization, NLC-polymer phase separation and NLC orientation proceed in balance under the action of the total field. At further temperature growth, these processes begin to compete with each other, and above 50 °C, the diffraction efficiency decrease significantly. The sensitivity of the material for hologram records is commonly characterized by the parameter (Ortler et al., 1989):

$$g = \eta^{1/2} / W,$$
 (14)

where *W* is the record energy density; η is the corresponding diffraction efficiency. For our samples, this parameter lies within the limits of 0.04 – 0.15 cm²/J, regarding the thickness, which is lower than in photo-reactive materials (0.2 – 0.5 cm²/J) (Ducharme et al., 1991; Eich & Wendorff, 1990).

At the electric field imposing, the NLC molecules are oriented along the field (since $\Delta \varepsilon > 0$), and for the radiation incident on the polarization holographic grating, the refractive index modulation is very small, and hence there is no radiation diffraction on such a grating.

Fig. 9 presents the optical response of the radiation diffracted on the polarization holographic grating, to the directing alternating electric field. It is evident that the condition of n = -n, satisfied for the director in the NLC, provides doubling frequency of the directing field by the optical response.

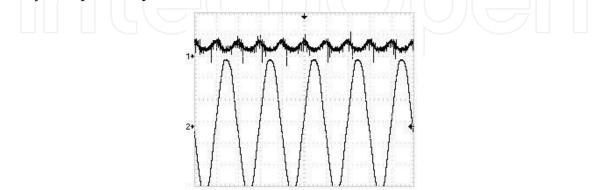


Fig. 9. Optical response of the polarization holographic grating (1) to the alternating voltage with frequency of 50 Hz (2).

The value of the electric field needed for the LC re-orientation depends on many factors: LCcomposite thickness, droplets size, optical and dielectrical anisotropy (Zharkova & Sonin, 1994).

Fig. 10 shows the dependence of the diffraction efficiency of the polarization holographic grating on the applied alternating electric voltage with the frequency of 50 Hz for various conditions of the grating formation. Evident that the dependence has a threshold nature, and the curve inclination is caused by the polidispersity of NLC droplets formed in the LC-composite.

Moreover it is seen that for the LC-composites of the same thickness, the mode of formation of the polarization holographic gratings influences the critical electric fields. The critical field is smaller for the gratings formed at lower energy density than for the gratings recorded at the higher energy density. It is associated with the effect of the radiation energy on the LC-composite morphology and in particular on the NLC droplets size, and the critical field is inversely proportional to the droplet size (Zharkova & Sonin, 1994).

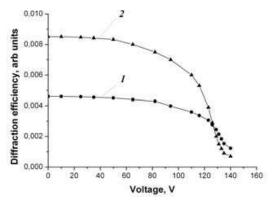


Fig. 10. Diffraction efficiency of the polarization holographic gratings vs the applied voltage. 1 – record energy density 1.2 J/cm^2 ; 2 – record energy density 0.45 J/cm^2 . LC-composite thickness is 10 µm. Spatial frequency of the grating is 53 mm⁻¹.

As follows from Figure 11, the critical field value highly depends on the LC-composite thickness, in which the polarization holographic grating forms. The critical electric field rises along with the thickness.

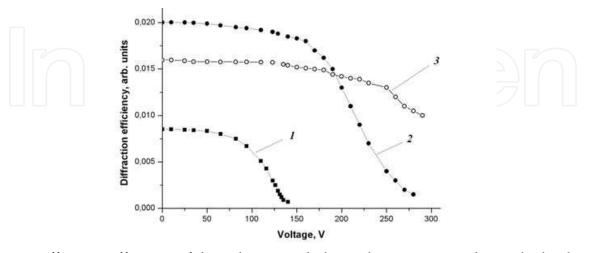


Fig. 11. Diffraction efficiency of the polarization holographic gratings vs the applied voltage for various thicknesses of the LC-composites. 1 – thickness of 10 μ m; 2 – 20 μ m; 3 – 50 μ m. Spatial frequency of the grating is 53 mm⁻¹.

It is also possible to control the diffracted radiation by means of the temperatures, since the physical properties of the NLC are sensitive to this parameter. In this context, we can observe the thermooptical switching into the state when the grating is "erased". Fig. 12 presents the dependence of the diffraction efficiency on the temperature.

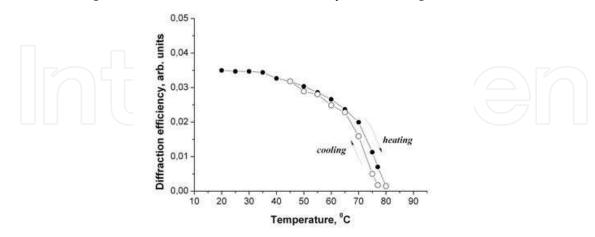


Fig. 12. Diffraction efficiency of the polarization holographic gratings vs the temperature. Sample thickness is 20 μm. Spatial frequency of the grating is 34 mm-1.

As is seen from the Figure, as the temperature grows, the diffraction efficiency decreases and at ~80 $^{\circ}$ C the diffraction properties of the grating disappear, i.e. the nematic liquid crystal transforms into the isotropic liquid. The temperature of the NLC – isotropic liquid transformation is considerably lower for the samples under study, than it is for the pure NLC used in the initial composition, which may result from the NLC pollution by the substances of the prepolymer composition, above all, by the polymer. At the grating cooling, as is seen from the Figure, its diffraction properties are recovered with a slight hysteresis.

Special structure of the NLC occurring at the polarization holographic gratings record causes the dependence of the diffraction efficiency on the incident radiation polarization and also the difference between the states of the diffracted and non-diffracted radiation.

In both cases, as the polarization grating is recorded by the *s*- and *p*-polarized beams, or it is recorded by the beams polarized at the angles of $\pm 45^{\circ}$, the intensities of the beams diffracted in the first order I_{+1} and I_{-1} were similar. Fig. 13 shows the dependence of the diffraction efficiency of the polarization holographic grating on the azimuth polarization angle of the probe beam α .

It is seen that the recorded gratings have the polarization selectivity, the diffraction efficiency highly depends on the probe beam polarization direction. At the incident s - or p – polarized probe beam on the grating, which is formed also by the s - and p – polarized beams, the non-diffracted beam (the 0 order) had the same polarization as the incident one, and the beams diffracted in the first order had the orthogonal polarization, i.e. if the incident beam had the s – polarization, the beams diffracted in the first order had the polarization and vice versa. If the sample was beamed by the radiation polarized at the angle of ±45°, the non-diffracted beam (the 0 order), as well as the beams diffracted in the first order in the same polarization of the light incident on the sample.

For the gratings formed by the beams polarized at the angles of $\pm 45^{\circ}$, with the probe incident beam polarized at the angle of $\pm 45^{\circ}$ or -45° , the beams diffracted in the first order had the orthogonal polarization, i.e. -45° and $\pm 45^{\circ}$, respectively. The non-diffracted beam (the 0 order) had the same polarization as the incident one. If an *s* - or *p* -polarized beam beamed such a

grating, the non-diffracted beam (the 0 order), as well as the beams diffracted in the first order, had the same polarization coinciding with the polarization of the light incident on the sample.

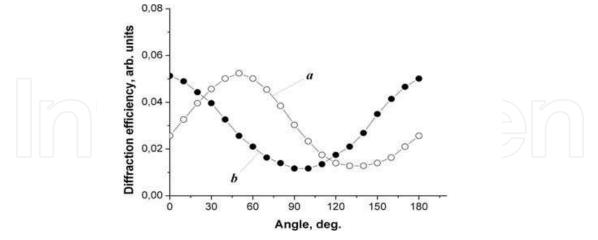


Fig. 13. Diffraction efficiency of the polarization holographic grating vs the azimuth angle of the probe beam polarization: a – the grating is recorded by the s- and p-polarized beams; b – the grating is recorded by the beams polarized at the angle of $\pm 45^{\circ}$ to the incident plane. Spatial frequency of the gratings is 34 mm⁻¹. Solid curves are the calculations of the diffraction efficiency by using Eq. (12). Sample thickness is 20 µm

The properties of the polarization gratings described above enable to control the diffracted radiation. Fig. 14 presents the photos of the diffraction patterns obtained at the diffraction of the s – polarized probe beam on the grating recorded by the s – and p – polarized beams. When the polarizer and analyzer are crossed (Fig. 3), the non-diffracted beam (the 0 order) can be suppressed, and when they are parallel, the same is valid for the beams diffracted in the first order.

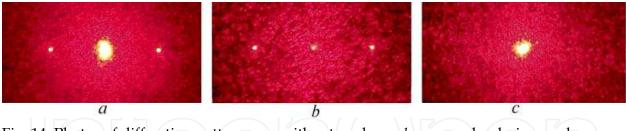


Fig. 14. Photos of diffraction patterns. *a* – without analyzer; *b* – crossed polarizer and analyzer; *c* – parallel polarizer and analyzer.

5. Conclusions

Thus, the one-stage process of record involving the polarization holography method results in the high-structured polarization holographic gratings with controllable supramolecular structure in the LC-composites. The diffraction efficiency of such gratings depends on the grating record energy, its formation temperature, NLC concentration in the prepolymer composition. It has been established experimentally that the gratings with the highest diffraction efficiency form at the temperatures of 30 - 45 °C, the NLC concentration in the prepolymer composition should be 35 - 38 mass %. The threshold record energy at which the maximal diffraction efficiency is reached is 2.5 J/cm² (for the grating up to 20 µm,

with the spatial frequency of 34 mm⁻¹). The total energy needed for the grating formation increases along with the spatial frequency of the recorded polarization grating, regardless its thickness.

The optical properties of the polarization holographic gratings can be controlled by the electric field; the doubling of the control voltage frequency by the optical response is guaranteed. The diffraction efficiency of such gratings and critical electric fields depends on the LC-composite thickness and record energy density of the grating. Increase of both parameters results in the critical field increase.

As the grating is heated up to \sim 80 °C, its diffraction properties disappear. The experiment has revealed the reduction of the NLC – isotropic liquid transformation temperature as compared to the pure NLC case, which may result from the NLC pollution by the substances of the prepolymer composition, above all, by the polymer. At the grating cooling, as is seen from the Figure, its diffraction properties are recovered with a slight hysteresis.

The diffraction efficiency of the polarized holographic gratings depends on the azimuth angle of the probe beam polarization. It has been established experimentally that the highest diffraction efficiency corresponds to the angle $\alpha = 45^{\circ}$ for the gratings formed by the s- and p-polarized beams and the angle of 0° in the case of the beams polarized at the angles of ± 45°. The possibility of "suppressing" of the beams diffracted on the polarization grating has been demonstrated.

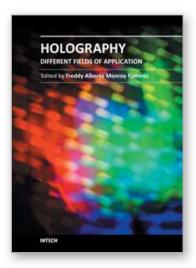
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This book depicts some differences from the typical scientific and technological literature on the theoretical study of holography and its applications. It offers topics that are not very commercial nor known, which will allow a different view of the field of optics. This is evident in chapters such as $\hat{a} \in \mathbb{C}$ Electron Holography of Magnetic Materials $\hat{a} \in \hat{a} \in \mathbb{C}$ Polarization Holographic Gratings in Polymer Dispersed Formed Liquid Crystals, and $\hat{a} \in \mathbb{C}$ Digital Holography: Computer-generated Holograms and Diffractive Optics in Scalar Diffraction Domain $\hat{a} \in \mathbb{C}$. The readers will gain a different view of the application areas of holography and the wide range of possible directions that can guide research in the fields of optics.

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