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CONTROLLED HOLOGRAPHIC RECORDING IN AZO-CONTAINING LIQUID-CRYSTAL POLYMER FILMS

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The dynamics of holographic recording processes in azo-containing side-chain liquid-crystal polymer films is experimentally studied. The possibility of effective control of optical recording conditions by applying an aligning electric field and also by varying the temperature of the polymer layer is demonstrated. In a polymer film 50 μm in thickness at a diffraction efficiency of around 75%, a sensitivity of around 21 cm^2/J is attained. High-density recording in the form of a Fourier-transform hologram is effected.

The possibility of controlling the diffraction characteristics of photosensitive media with liquid-crystal properties is of considerable interest in problems of information optical processing and storage. In the works known to date [1–3], this possibility, however, has been considered only at the stage of reading of pre-recorded holograms. At the same time, the optical characteristics of liquid-crystal polymer materials containing azo dye fragments can be controlled by means of an electric field in the course of holographic recording. The reason is that the predominant orientation of the electrically anisotropic azo chromophores participating in the formation of the liquid-crystal mesophase coincides with the direction of the liquid-crystal director of the polymer. The efficiency of photoprocesses is governed by the projection of the major axis of the dye on the direction of the electric field vector of the light wave and depends on the director orientation. Under the effect of the external aligning electric field applied there takes place the reorientation of the liquid-crystal director in the polymer layer, which changes the efficiency of interaction between the azo dye contained in the polymer and light and finally leads to a change in the light-induced refractive index Δn_{ind}, as well as in the effective holographic recording depth [4].

This work presents the results of experimental studies into the dynamics of holographic recording processes in azo-containing liquid-crystal polymer films. Considered here is the effect of the external aligning electric field applied and of the optical recording conditions, such as the temperature of the sample and the intensity and polarization of the recording beams.

The experiments were carried out using film samples of Grade SPK4/4 comb-shaped liquid-crystal polymer containing 20 mol.% of chemically bound azo dye fragments [5]. The polymer is characterized by a high energy of interaction between chromophores and nonphotosensitive polar groups (cyano biphenyl fragments) and forms a nematic phase in the temperature range 40–140 °C. At temperatures below 40 °C, the polymer is in a vitreous state. The optical nonlinearity of the compound is associated with the reversible *trans-cis* photoisomerization followed by molecular reorientation. Note that azo-containing polymers of the structure of interest possess a high sensitivity in the blue-green region of the spectrum, high stability of parameters, and high resistance against repeated exposures to light [5].

The experiments were performed with a sample cell (Fig. 1) consisting of polymer film *5* with a thickness of $L = 50$ μm confined between glass plates *1* and *3* coated with transparent current conducting In_2O_2 layers. The polymer layer was preliminarily oriented by rubbing the surfaces of the plates. As a result, they attained an initial (nonrigid planar) orientation of mesogene polymer fragments. An alternating voltage of $U_a = 10$–100 V was applied across the conductive coatings of plates *1* and *3*, which made it possible to orient the liquid crystal director of the polymer along the field (as a result of the turning of the mesogene fragments). To exclude undesirable hydrodynamic effects, the applied a. c. voltage frequency was around 350 Hz. The preliminary orientation of the polymer film prevented the formation of a polydomain structure when the electric field was removed. The heating of the polymer film was effected by passing electric current through the conductive coating of plate *3*. The sample temperature was measured by means of semiconductor transducer *4* and stabilized by digital control circuit *7*. The working aperture of the sample cell amounted to some 4 cm^2.

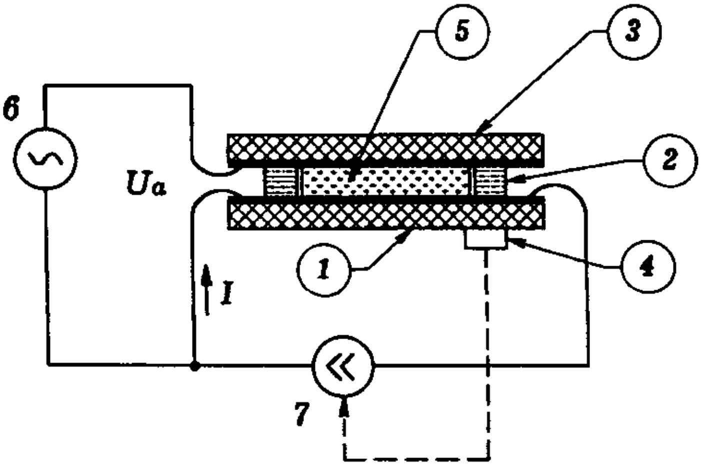


Fig. 1

Schematic and circuit diagrams of the polymer sample cell: (*1*) glass plate with a heating coating, (*2*) spacers, (*3*) glass plate with a conductive coating, (*4*) temperature transducer, (*5*) polymer film, (*6*) a. c. generator, (*7*) source of the heating current *I*.

Holograms were recorded in the polymer sample "in transmitted light" by the interferometric method using the second-harmonic radiation of an LTN402A YAG:Nd laser ($\lambda \cong 532$ nm). The optical densities of the film at $\lambda \cong 532$ nm were $D_{\|} \cong 2.8$ along and $D_{\perp} \cong 0.9$ across the predominant orientation of the liquid crystal director. The experimental setup is schematically shown in Fig. 2. The output laser radiation passes through electromechanical shutter *2* and collimator *4* with a diaphragm some 20 μm in diameter and hits splitting cube *6*. Ancillary mirrors *1*, *3*, and *5* are used to reduce the setup overall dimensions. The light beams obtained are reflected by mirrors *11* and *12* and form an interference pattern in the plane of polymer film *18*. The polarizations of the beams are set by double Fresnel rhombs *7* and *10*, and their intensities are equalized by means of neutral filter *13*. When recording holograms of amplitude masks (images), use is made of elements placed in the object beam path: electromechanical shutter *8*, amplitude image transparency *9*, and objective lens *14* with a focal length of F. Transparency *9* is placed at a distance of $2F$ from objective lens *14*, and polymer sample cell *18* is in the focal plane of the lens. With this geometry, there takes place the recording of the hologram of the spatial frequency spectrum of the image (a Fourier-transform hologram). In the experiment, the fringe frequency corresponding to an angle of convergence of the recording light beams of $2\theta \cong 28$ deg is $\Lambda^{-1} \cong 909$ lines/mm. The reading of holograms is effected by radiation from a He–Ne laser ($\lambda_P \cong 633$ nm, $I_P \cong 20$ mW/cm^2) simultaneously with their recording. The optical densities of the polymer film at $\lambda_P \cong 633$ nm are $D_{\perp} \cong 0.07$ and $D_{\|} \cong 0.2$. The light intensity I_B in the Bragg diffraction order is measured with phototransducer *15*, and, in addition, the polarization of the transmitted radiation is analyzed in the zero order by means of a system consisting of polarizing cube *19* and phototransducers *16* and *17*. To record the image in reading the hologram recorded (in experiments with images), use is made of CCD camera *21* and additional objective lens *20*. The general control of the experimental setup and data acquisition from the transducers are effected by a personal computer equipped with a ADC/DAC board.

In the experiments, we studied the dynamics of the diffraction efficiency, namely, $\eta(t) = I_B/I_P$ (I_P is

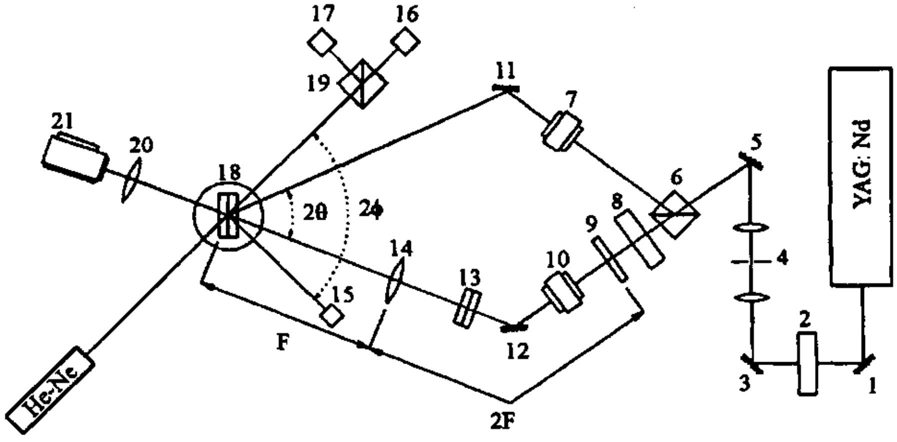


Fig. 2

Schematic diagram of the experimental setup (for explanation, see text).

the intensity of the incident probe beam), at various temperatures of the polymer film, various intensities of the recording beams, and various strengths of the applied electric field. Figure 3*a*, *b* presents time dependences of the diffraction efficiencies $\eta(t)$ for various effective values of the applied voltage U_a. The curves of Fig. 3*a* were obtained in the case of like polarizations (S–S) of the recording beams. The intensities of the beams were $I_{w1} \cong I_{w2} \cong 85$ mW/cm^2, and the temperature of the polymer, $T = 70\,^\circ$C. Similar dependences for the case of orthogonal polarizations (S–P) are presented in Fig. 3*b*. As can be seen from these figures, raising the voltage U_a for the S–S-polarized beams leads to a noticeable growth of the maximum diffraction efficiency ($\eta_{\max}$), whereas that in the case of S–P-polarized beams causes $\eta_{\max}$ to drop. Such a behavior points to the homeotropic ordering of azo chromophores in the polymer film under the effect of the applied electric field and also to a strong anisotropy of their form. As the applied voltage is raised ($U_a > 40$ V), the maximum diffraction efficiency $\eta_{\max}$ rapidly reaches its ultimate value ($\sim 77\%$), whereas in the region $U_a < 10$ V, the quantity $\eta_{\max}$ is practically independent of the applied voltage. This means that the process of reorientation of the liquid crystal director in the polymer layer under the effect of the applied field is of a threshold character, with saturation at high voltages.

Varying the intensities of the recording beams and the polymer temperature makes it possible to materially affect the dynamic characteristics of the recording process. The behavior of $\eta(t)$ at various intensities of the light beams is illustrated in Fig. 3*c*. The layer temperature was $T \cong 70\,^\circ$C, the recording beams were S–S-polarized, and the applied voltage $U_a \cong 80$ V. As one can see, the growth dynamics of the diffraction efficiency is nonlinear in exposure time: the time it takes for the diffraction efficiency to reach its maximum value $\eta_{\max}$ decreases in a nonlinear fashion as the beam intensities are increased. Such a behavior can be explained by a fast rise of nonlinear absorption in the polymer surface layers, leading to a reduction of the effective holographic recording depth. The temperature dependences $\eta(t)$ are presented in Fig. 3*d*. In this experiment, the recording beams were S–S-polarized and their intensities were $I_{w1} \cong I_{w2} \cong 85$ mW/cm^2. An aligning voltage of $U_a \cong 80$ V was applied across the polymer film. The maximum holographic sensitivity $S = \sqrt{\eta_{\max}}/(I_{\text{tot}} t_{\max}) \cong 21$ cm^2/J (I_{tot} is the total intensity of the recording films and $t_{\max}$ is the time needed for the diffraction efficiency to reach its maximum value $\eta_{\max}$) was attained at $T \cong 75.7\,^\circ$C (curve *4*). It follows from Fig. 3*d* that at temperatures close to $T_c \cong 78\,^\circ$C there takes place a sharp reduction of $\eta_{\max}$ but the rate at which $\eta_{\max}$ is reached is also increased in that case. Such a behavior is characteristic of phase transitions. In actual fact, in polymer films at a temperature of T_c which is much below the nematic-to-isotropic transition for bulk specimens ($T_{NI} \cong 104\,^\circ$C), there is observed a spontaneous homeotropic ordering of the polymer molecules similar to a first-order transition. The polymer temperature dependences of the variation rates of the light-induced refractive index (dn_{ind}/dt) in the course of recording and relaxation are presented in Fig. 4 (curves *1* and *2*, respectively). These curves can be approximated well enough by functions of the form $dn_{\text{ind}}/dt \sim (T - T_c)^{-\gamma}$. Here $\gamma \cong 0.7$ for the

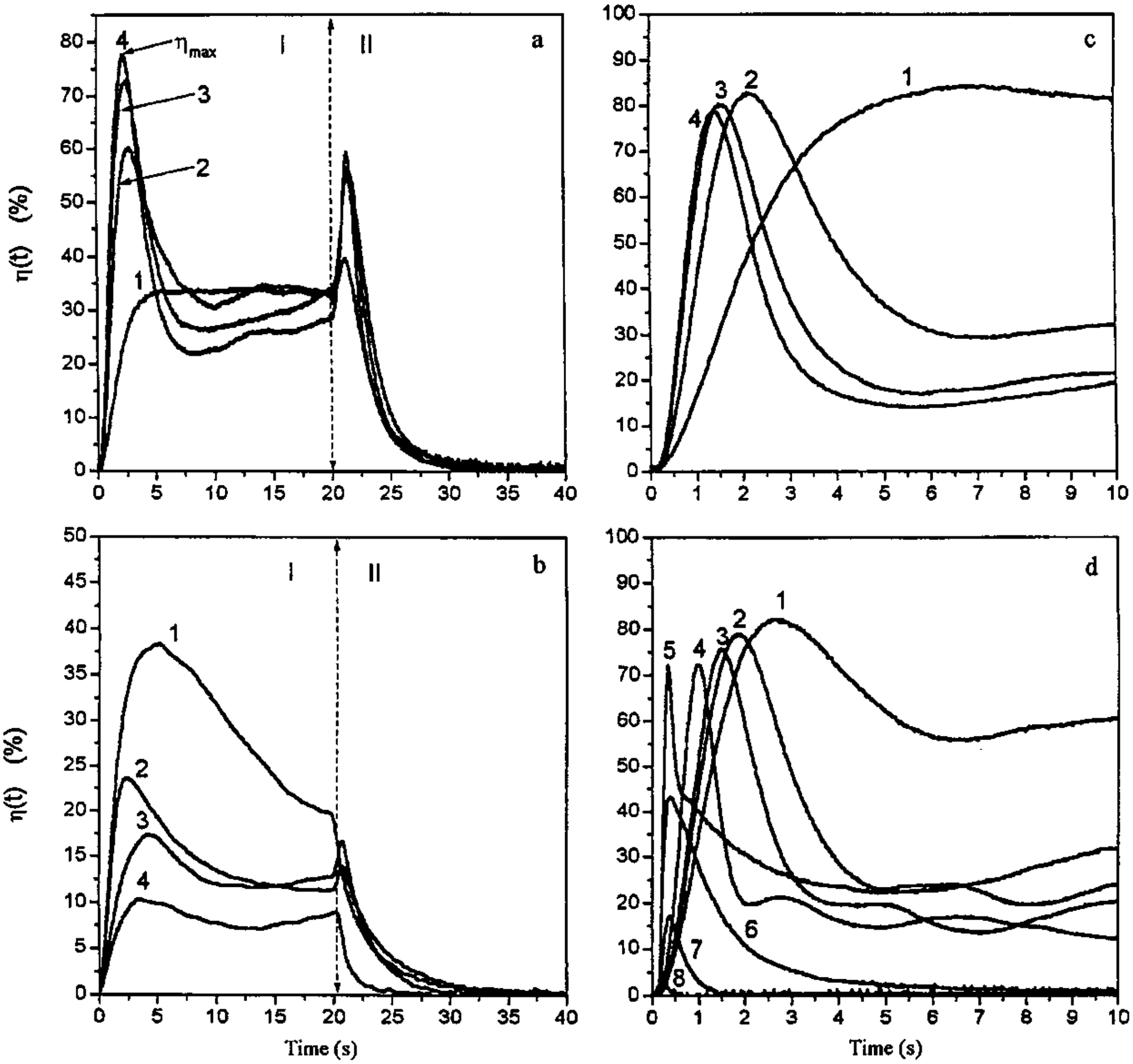


Fig. 3

Dynamics of the hologram diffraction efficiency in a polymer film in an aligning electric field. (*I*) hologram recording, (*II*) relaxation (recording beams are shuttered) in (*a*) parallel and (*b*) orthogonal polarizations of the recording beams: (*1*) $U_a = 10$ V, (*2*) $U_a = 20$ V, (*3*) $U_a = 40$ V, (*4*) $U_a = 80$ V. (*c*) Recording at various intensities of the recording beams: (*1*) 48 mW/cm^2, (*2*) 85 mW/cm^2, (*3*) 122 mW/cm^2, (*4*) 192 mW/cm^2. (*d*) Recording at various polymer temperatures: (*1*) 69.7 °C, (*2*) 70.8 °C, (*3*) 71.6 °C, (*4*) 73.6 °C, (*5*) 75.7 °C, (*6*) 76 °C, (*7*) 76.2 °C, (*8*) 76.5 °C.

process of recording and $\gamma \cong 0.5$ for that of relaxation.

The experiments on recording the Fourier-transform holograms of amplitude masks were conducted using a standard test object (Fig. 5). Holograms were recorded with shutters *2* and *8* open (see Fig. 2). The optimum exposure time was $t_0 \cong 0.5$ s at a sample temperature of $T \cong 65$ °C and the object and reference beam powers of $P_{w1} \cong P_{w2} \cong 0.9$ mW. Once the recording ended, shutter *8* was closed and the remaining reference beam performed the reading of the hologram. The image was recorded in the Bragg diffraction order by CCD camera *21*. As seen from Fig. 5, applying an electric field in the course of holographic recording

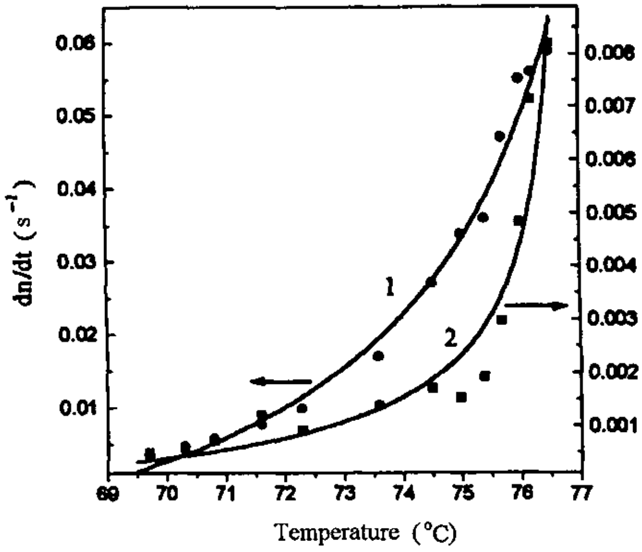
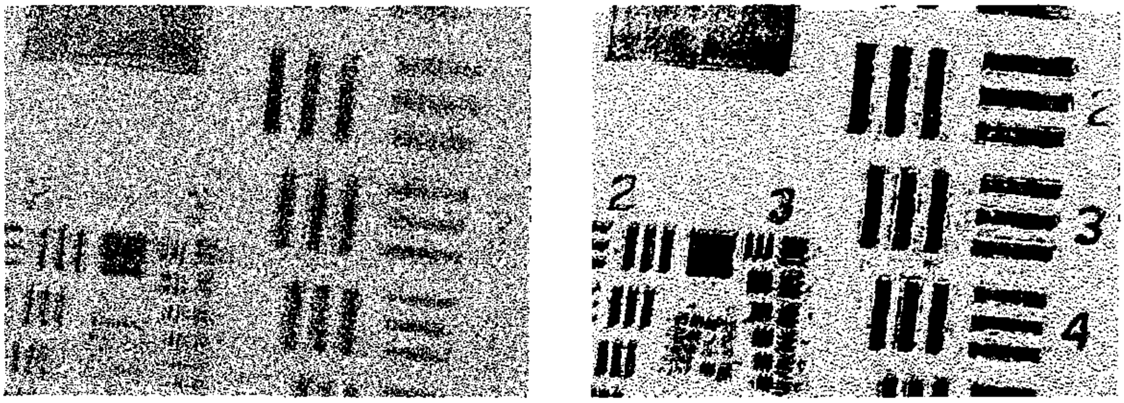


Fig. 4

Temperature dependence of the variation rate of the light-induced refractive index in (*1*) recording and (*2*) relaxation.

leads to a substantial growth of the signal intensity, which is associated with the growth of the diffraction efficiency. To eliminate saturation of Δn_{ind} in the region of zero spatial frequency of the hologram (occurring as a result of high light intensity), the plane of the polymer film was somewhat displaced from the focal plane of objective lens *14*. The size of the recording region was in that case $d \cong 200$ μm.



a

b

Fig. 5

Fragments of images of Fourier-transform holograms read: (*a*) recording without any electric field applied; (*b*) recording in a field of $\sim 1.2 \times 10^4$ V/cm.

The minimal resolvable element of the test object in Fig. 5*b* corresponds to a fringe frequency of around 11 lines/mm, and a complete image fragment contains $N \cong 300 \times 200$ elements (information bits). With these parameters being as they are, the information capacity of the hologram is $q = 4N/\pi d^2 \cong 1.9 \times 10^6$ bits/mm^2 (or some 24 MB/cm^2). Inasmuch as the holographic recording in the polymer film takes place in the spatial (Bragg) mode, a higher recording density can be attained by using angular selection [6].

The effect of the applied electric field on the holographic recording efficiency can be explained by molecular reorientation processes. The application of an electric field to a liquid-crystal polymer film causes the liquid crystal director to align with the field. In that case, the proportion of homotropically oriented chromophores increases, which leads to a reduction of light absorption (an increase of the recording depth) and also to a decrease of the chromophore photoisomerization efficiency in the film near-surface layers. However, due to the greater recording depth, the diffraction of light takes place on a holographic grating having a greater extension in the medium (Bragg diffraction conditions), and it is this fact that provides for the resultant growth of the diffraction efficiency [7]. Note that in our case the possibility to change the diffraction conditions is due to the high light absorption by the polymer at $\lambda = 532$ nm along the liquid crystal director, $\beta_{||} \geq 1/L$, and the great absorption anisotropy, $\Delta\beta = \beta_{||} - \beta_{\perp}$ (for the sample used, $\beta_{||} \cong 560$ cm^{-1} and $\beta_{\perp} \cong 180$ cm^{-1}, film thickness $L = 50$ μm).

To conclude, let us formulate the main results of this work. The dynamics of holographic recording in azo-containing liquid-crystal polymer films is experimentally investigated. It is demonstrated that the application of a homeotropically aligning electric field ($\sim 10^4$ V/cm) leads to a substantial growth (by more than twice) of the maximum attainable diffraction efficiency. The temperature and exposure dependences of the holographic recording dynamics are obtained. The existence of optimal (from the standpoint of diffraction efficiency and exposure time) dynamic hologram recording conditions is discovered. The possibility of high-density recording of images is demonstrated.

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