Optical data recording by laser pulses in liquid-crystal cells with an azo-modified surface

S V Serak, A V Agashkov, V Yu Reshetnyak

Abstract. **The effect of trans–cis photoisomerisation of azo-fragments of a polymer film on the molecular reorientation of a liquid crystal is studied. It is shown that, using nanosecond laser pulses, one can perform both the reversible and static data recording in liquid-crystal cells with an azo-modified surface. The rise time of the reorientation is measured by the methods of dynamic holography to be about ~ 30 μs, and the grating efficiency achieves 15 %.**

1. Introduction

At present, liquid crystals (LCs) find wide applications not only as materials for displays but also as nonlinear optical media in devices for optical data recording and storage, in optical modulators, and laser elements such as cavity *Q* switches, phase-conjugate mirrors, wavelength-tunable mirrors, wave plates, etc. One of the urgent problems is a search for new materials and methods for controlling the orientation of a LC matrix using laser radiation. For this purpose, photosensitive orienting materials are employed, in particular, azo polymers, which exhibit a high nonlinear response [1 – 8].

By irradiating the orienting surface of an azo polymer containing structural fragments capable of *trans–cis* photo-isomerisation by polarised laser light, one can efficiently perform the molecular reorientation of the LC and achieve a maximum change in the refractive index of the LC layer from n_e to n_o. In this case, the twist-deformation of the director can change to the planar one [2] or the planar deformation can change to the homeotropous one [3], etc. The type of transformation depends on the chemical structure of azo-containing materials, the cohesion energy of LC molecules with a surface, and the parameters of laser radiation (polarisation, intensity, and the irradiation duration). In such LC devices, stable holograms with a memory and resolution of no worse than 100 lines/mm can be recorded by radiation from a pulsed Ar laser with the energy density of about 1 J cm^{-2} [2].

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Another method for controlling the director of the LC layer is the induction of the easy-orientation axis on an unoriented polymer surface by laser radiation [4, 9 – 10]. In this case, molecular photoisomerisation in the LC volume doped with dyes and adsorption of isomers on the unoriented surface play the role. The specific feature of this method is that the layer director is rotated in the plane of the LC cell substrate, whereas in a dc electric field the molecules are rotated perpendicular to substrates. In addition, this method allows the polarisation of the output radiation to be controlled. The authors of Ref. [9] reported a high spatial resolution of gratings (up to 200 lines/mm) recorded in such materials at the energy density of about ~ 0.1 J cm^{-2}.

At present, the mechanism of photoinduced orientation in polymer films containing azo-chromophore groups (the $-N=N-$ bond) is well studied [11, 12]. It includes two stages. At the first stage, an anisotropic depletion of the angular distribution of *trans*-isomers takes place due to the accumulation of *cis*-isomers along the polarisation direction. At the second stage, molecules are rotated during the *trans–cis–trans*-isomerisation cycle [11].

In most cases, the molecular fragment $-N=N-$ is oriented perpendicular to the field upon the interaction of azo dyes with polarised laser radiation, and the layer director gradually deviates from the polarisation direction [13]. The authors of most papers have studied the reorientation of LCs upon steady-state *trans–cis–trans*-photoisomerisation in polymer films induced by radiation from cw gas lasers. The rise time of reorientation was quite large – several minutes. An increase in the response speed of this effect and a search for media with a large nonlinear response are urgent problems both for practical applications and scientific studies.

The mechanism of *trans–cis*-isomerisation is a nonlinear mechanism, which can be initiated in the field of short laser pulses because the time of conformation transformations of molecules is a few picoseconds [14]. Earlier, we induced for the first time the isothermal phase transition (disordering of the LC layer due to the accumulation of *cis*-isomers in it) in LC azobenzenes by single radiation pulses of the second harmonic from a ruby laser [15]. Further studies [16] revealed the reorientation of the director in these media under the same excitation conditions. The study of the dynamics of reorientation relaxation upon excitation by intense short laser pulses allows one to establish the stage at which collective rotational motion of LC molecules arises.

In this paper, we study the possibility of data recording by nanosecond pulses of the second harmonic of a Nd:YAG

laser in LC cells with an azo polymer film deposited onto one (input) substrate. The dynamics of molecular reorientation of LCs is investigated upon irradiation by one or two laser beams using dynamic holography and polarisation microscopy. The surface cohesion energy on the input substrate is theoretically estimated before and after irradiation and its variation during irradiation is calculated.

2. Experimental description

2.1. LC samples

We prepared LC cells in which one of the substrates (on the side of an incident pump beam) was covered by azo polymer, while the second substrate (output) was covered by a rigidly rubbed orienting polyvinyl alcohol (PVA) film. Poly-(disperse red-1)-methacrylate-co-methyl methacrylate (DR1-PMMA) copolymer was used as an azo polymer. Molecules of a 4-diethylamine-4′-nitroazobenzene azo dye [(DR1), the weight concentration of 24 %] were chemically bonded to the main polymer chain. The structural formula of the azo polymer and absorption spectra are presented in Fig. 1.
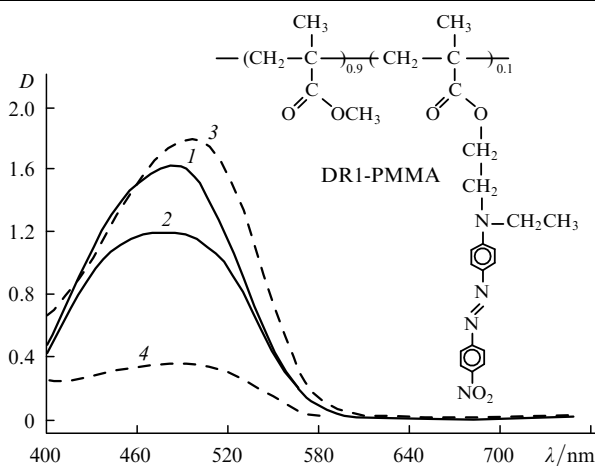

Figure 1. Structural formula and absorption spectra of azo polymer DR1-PMMA (1, 2) and azo dye DMANAB in a planar layer of 5CB (3, 4) for orientations $E \parallel q$ (1), $E \perp q$ (2), $E \parallel L$ (3), and $E \perp L$ (4).

The absorption spectrum of the film has a maximum at 485 nm. The azo polymer solution in methylene chloride (3 %) was deposited onto a glass substrate by pulling the substrate from the solution in one direction. The directions of azo polymer pulling (q) and PVA film rubbing (n) were mutually perpendicular. The absorption of the azo polymer film exhibits weak dichroism $d = (k_{\parallel} - k_{\perp})/(k_{\parallel} + k_{\perp}) = 0.15$ caused by the method of film preparation. Here, $k_{\parallel}$ and $k_{\perp}$ are the absorption coefficients, which correspond to the mutually parallel and orthogonal orientations of the polarisation vector E of the light wave and the direction q.

After prolonged irradiation of the LC cell by cw radiation from a He–Cd laser at 442 nm, the cell was 'reddened' and the absorption spectrum shifted to the red by 10 nm, which confirms the formation of cis-isomers [12]. We used in our experiments a 4-pentyl-4′-cyanobiphenyl (5CB) nematic. The thickness of the LC layer was 10 μm.

In the LC layer with an unoriented azo-polymer surface, a stable orientation arises, which can be planar, homeo-

tropous or twisted [17]. We studied the orientation of the LC cell using a polarisation microscope and by measuring dichroism of absorption of the LC layer doped with a dye whose molecules were elongated and isomorphous to the LC molecules [18].

To do this, we added to the 5CB nematic a small amount (0.1 %) of a 4-dimethylamino-4′-nitroazobenzene (DMA-NAB) azo dye. The order parameter of DMANAB in the planar 5CB layer oriented by rubbed PVA films on two substrates was $S = (k_{\parallel} - k_{\perp})/(k_{\parallel} + 2k_{\perp}) = 0.6$ and was approximately equal to that of the LC layer.

The absorption coefficients $k_{\parallel}$ and $k_{\perp}$ of the LC layer correspond to orientations $E \parallel L$ and $E \perp L$, where L is the direction of the LC director. In this case, the absorption dichroism of the layer is $d = 0.68$. Immediately after the filling of the LC cell with the azo-modified surface the absorption dichroism was $d = 0.43$, which is lower than in a planar cell. The study of samples with a polarisation microscope showed that they had the orientation that was close to a planar one, with molecules oriented at an angle to the input substrate. After one–two hours, the absorption dichroism dropped to 0.2, corresponding to the establishment of another, more stable orientation. A twist structure appeared with a twist angle φ' on the input substrate, the angle being dependent on the azo polymer film thickness (which varied from 0.2 to 0.5 μm) and the method of the film deposition.

2.2. Photoinduced reorientation of the director in LC cells with an azo-containing polymer surface

We studied orientation variations in the volume of an undoped 5CB layer photoinduced by the azo-modified surface. The scheme of the experiment is shown in Fig. 2. Nonlinear optical properties of LC cells were studied both in crossed polaroids 1 and 2 (screen 6 blocks one of the exciting beams) and by the method of dynamical gratings (the screen was absent). The intensity of a probe beam from a 633-nm, 1-mW He–Ne laser that passed through the centre of the excitation region of LC cell 5 was measured with photodetector 7. Excitation was performed by the second harmonic of a Nd:YAG laser at 532 nm. The energy of the laser pulse was 1 mJ, the energy of the amplified pulse was 5.5 mJ, and its duration was 10 ns.
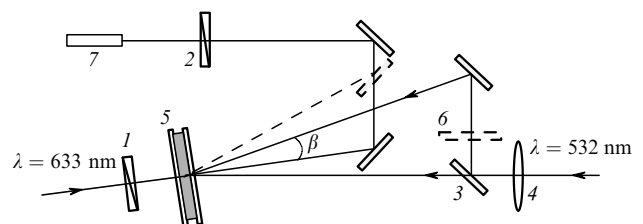

Figure 2. Schematic of the experimental setup for the study of the reorientation of a LC cell with the azo-modified surface produced by irradiation by the second harmonic from a Nd:YAG laser: (1) polariser; (2) analyser; (3) mirror, $R = 50\%$; (4) lens, $f = 1.5$ m; (5) LC cell; (6) screen; (7) photodetector.

The geometry of the experiment allowed us to study both the transmission kinetics and the rotation of the layer director in accordance with the Mauguin regime. We studied the LC cell with an azo polymer film on the input substrate whose orientation axis makes the angle φ' with the direction n of rubbing on the second substrate. In this cell illuminated

by polarised radiation from a Nd:YAG laser, the region appears that has the twist orientation different from the initial one.

We studied a change in the transmission of the probe beam that passed through a cell with an azo-modified film, which was placed between polaroids so that the rubbing direction n of the output surface coincided with the direction of the axis of polariser *1*, while the axis of analyser *2* was turned by the angle $90° + \varphi'$, which provided the minimum transmission in the cell. In such a geometry, the polarisation vector of radiation from a He–Ne laser follows the director rotation according to the Mauguin regime, and we can measure the rotation angle with the help of the analyser. When the polarisation vector E_{Nd} of radiation from a Nd:YAG laser is parallel to n, the twisting angle φ of the director increases ($\varphi > \varphi'$), and when $E_{Nd} \perp n$, the twisting angle decreases ($\varphi < \varphi'$). In other words, LC molecules are aligned perpendicular to the direction of radiation polarisation. The twisting angle depends on the excitation power.

One can obtain almost complete twist or planar reorientation, i.e., observe the untwisting of the layer director to $\varphi = 0$ or its additional twisting to $\varphi = 90°$. Fig. 3 shows the dependences of the twisting angle $\varphi = \varphi' + \Delta\varphi$ on the pump power density W and the time after irradiation. The twisting angle in Fig. 3a was measured 5 s after irradiation (the
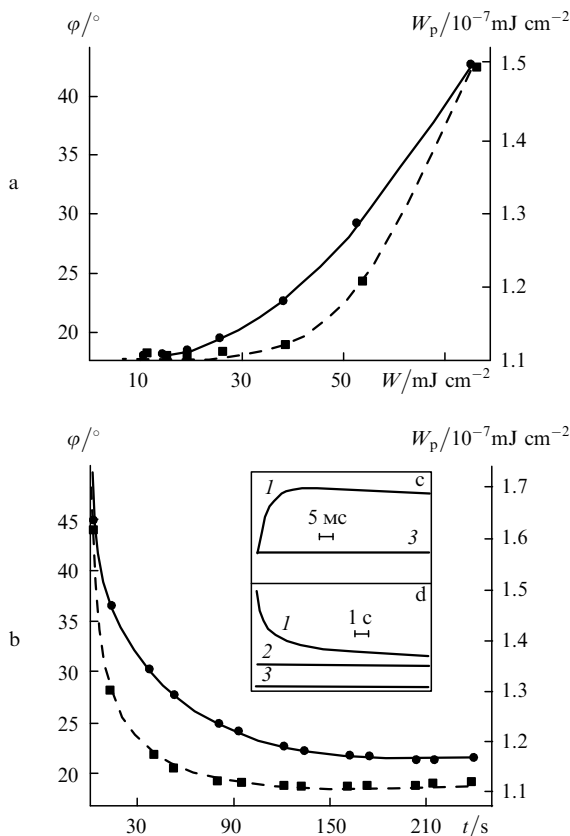
initial twisting angle was $\varphi' = 18°$). The nonlinear response and the time of the reorientation establishment substantially depend on the excitation intensity. There exists a threshold pump power density $W_{th} = 30$ mJ cm^{-2} from which the reorientation efficiently develops (Fig. 3a). One can see from the oscilloscope trace in Fig. 3a that the reorientation develops for ~ 10 ms.

Upon illumination by a single pulse with a power density of 80 mJ cm^{-2}, the decay occurs in two stages: first, the cell transmission rapidly decreases for ~ 1 s (Fig. 3d, curve *1*) and then it returns slowly for several minutes to its initial value. Excitation by two-three pulses causes the static recording of photoorientation, which persists for several hours. The slow decay of the induced twisting angle could be observed in a polarisation microscope.

Fig. 4 demonstrates the accumulation of photoinduced reorientation in a cell irradiated by a train of 5-mJ pulses with a repetition rate of 1 Hz. In the LC cell with the director initially twisted by 18°, the twisting angle increased to 38–70° after irradiation, depending on the number of pulses (Fig. 4a). Fig. 4b shows the oscilloscope trace of the reorientation dynamics in this case. One can clearly see an increase in the constant component of the pulse train. After irradiation by the first pulse, the initial orientation of the cell does not recover and the effect exhibits the tendency to accumulation upon irradiation by subsequent pulses and saturates after the seven pulse. The eight pulse performs the static recording of a spot (Fig. 4c), which persists for several months.
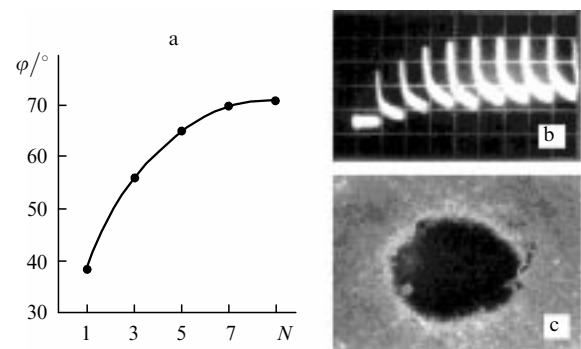

Figure 3. Dependences of the rotation angle φ of the director in a LC cell with the azo polymer surface (solid curve) and the energy of cohesion W_p with a surface (dashed curve) on the excitation energy density (a) and time after irradiation (b), and the time dependences of transmission of the LC cell on different time scales (c, d) immediately after the sweep start by a single pulse (*1*), after 30 s (*2*), and before the pulse action (*3*).

Figure 4. Accumulation of the photoinduced reorientation of a LC cell upon excitation by nanosecond pulses with an energy density of 80 mJ cm^{-2} and a repetition rate of 1 Hz (a, b), and the static recording of a spot by a train of 8 pulses (c)

The special feature of this type of recording is the formation of differently oriented structures at the centre and edges of the spot. After irradiation by a Gaussian beam with the polarisation vector directed along the rubbing direction n, the central region of a Gaussian spot proves to be 'twisted' to 63°, whereas the external region is 'untwisted' to zero. When the rotation angle of the orientation axis on the input substrate is close to 45°, the orientation forces interacting in the volume and on the surface can be distributed so that the spot will contain regions with different orientations. The pattern has the form of a four-leaf structure, where the twisted region is followed by the untwisted region. In this case, 'a cross' is detected in the probe beam, whose axes are fixed with the axes of crossed polaroids.

2.3. Dynamic and static gratings in LC cells with the azo polymer surface

We recorded both dynamic and static gratings in LC cells with the azo-modified surface by the interference radiation field of a Nd:YAG laser. Figs. 5a–d show oscilloscope traces of relaxation curves for dynamic gratings of different types. Such gratings are detected in several photoinduced exponentially decaying processes with the characteristic times equal to 5 μs (Fig. 5a), 3 ms (Fig. 5c), and 0.3 s (Fig. 5d). In the LC cell with the azo-modified surface, the azo polymer film and the near-surface LC layer can be heated by laser pulses, the LC molecules can be reoriented inside the layer, and the polymer surface can undergo photoinduced modifications. This will result in the formation of phase and amplitude gratings.
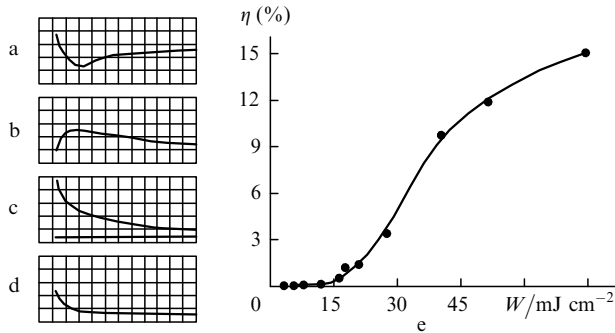

Figure 5. Relaxation of dynamic gratings of different types in a LC cell with the azo polymer surface (a–d) at the excitation energy density of 60 mJ cm^{-2}, and the dependence of the diffraction efficiency η of the orientation grating on the excitation energy density W (e). The time scale division for oscilloscopes traces is 5 μs (a), 50 μs (b), 5 ms (c), and 0.2 s (d).

The decay constants of the thermal and orientation gratings can be estimated from expressions [19]

$$\tau_t = \frac{\Lambda^2}{4\pi^2 D_t}, \quad \tau_\theta = \frac{\gamma \Lambda^2}{4\pi^2 K_{22}},$$

where D_t is the thermal diffusion coefficient; γ is the rotational viscosity of the LC; K_{22} is the elasticity constant for the twist-deformation of the director. For 5CB, $D_t \approx 1.8 \times 10^{-3}$ cm^2 s^{-1} [19], $\gamma \approx 10^{-2}$ N s m^{-2}, $K_{22} \approx 4.0 \times 10^{-12}$ N [20]. Then, for the grating period $\Lambda = 10$ μm (this period provides the maximum diffraction efficiency of the orientation grating), we obtain $\tau_t = 14$ μs and $\tau_\theta = 6$ ms. In cells with azo polymer films, the experimental time is close to the relaxation time τ_t of the thermal grating.

After thermal relaxation, the grating efficiency does not decrease but on the contrary increases for ~ 30 μs (the oscilloscope trace in Fig. 5b). We assume that this is caused by the time lag in the response of LC molecules to a change in the orienting properties of azo polymer, because other gratings, namely, the amplitude 'trans-cis' grating on the surface and thermal grating are formed during the action of a single pulse and decay after its termination. The relaxation time $\tau_\theta = 3$ ms is typical for the volume reorientation of LCs [16]. The slow relaxation with the decay time $\tau_s = 0.3$ s can be related to the photoinduced reorganisation of the polymer film. The study of kinetics of grating formation showed that when the irradiation intensity exceeded 70 mJ cm^{-2},

the brightness of diffraction maxima increased for 1 s and then a stationary state was established.

Fig. 5e shows the change in the diffraction efficiency of the orientation grating with increasing energy density of the pump pulse. The diffraction efficiency η was defined as a ratio of the maximum amplitude of the relaxation pulse of the orientation grating (the oscilloscope trace in Fig. 5b) for the first diffraction order of the probe beam to the amplitude of a constant signal on an oscilloscope corresponding to the intensity of the incident probe beam. The efficiency η reached its maximum value equal to 13 % for the grating period $\Lambda = 10$ μm. The corresponding change Δn in the refractive index can be estimated from the relation $\eta \sim (\Delta n \pi L / \lambda)^2$. It equals ~ 0.008 and characterises a high nonlinear response of the medium.

Using a single pulse with the radiation energy density no less than 80 mJ cm^{-2}, we recorded static gratings with the spatial frequency of no worse than 100 lines/mm, whose microphotographs is presented in Fig. 6. The relaxation time of the gratings achieved several weeks. Fig. 6b shows gratings recorded in a cell with the initial twisting angle equal to $42°$ against the background of regions with competing twisted and untwisted structures in the same spot.
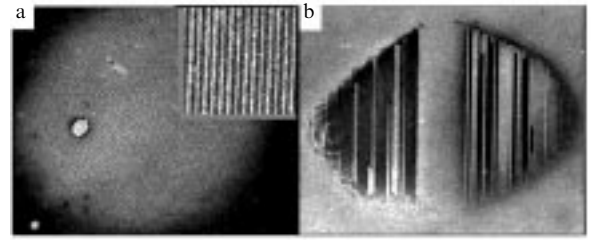

Figure 6. Microphotographs of gratings with a spatial frequency of ~ 100 lines/mm for the initial twist angle $\varphi' = 18$ (a) and $42°$ (b).

3. Theoretical estimates and discussion

The molecular reorientation of a LC is caused by the photoisomerisation of the dye chemically bonded to polymer chains of the azo polymer deposited onto one of the substrates of the LC cell. The rotation angle of the director depends on the laser radiation intensity and the direction of its polarisation, as well as on the distribution of elastic forces in the volume and on the surface cohesion. Knowing the rotation angle of the director on the input surface, we can estimate the surface cohesion energy and its variation during irradiation.

Consider a LC cell of thickness L in which the output substrate is rubbed and the director has an infinitely strong cohesion with it. The cohesion of the director with the input substrate is finite and the direction of the easy axis makes the angle φ_0 with the direction of rubbing $\boldsymbol{n}$ on the output substrate. Due to the symmetry of the problem, we will describe the director field by the angle $\varphi(z)$

$$\boldsymbol{n}(z) = \{\cos\varphi(z), \sin\varphi(z), 0\}, \tag{1}$$

where the z-axis is directed perpendicular to cell substrates.

The free energy functional has the form

$$F = \frac{1}{2}\int_0^L K_{22}\left(\frac{\mathrm{d}\varphi}{\mathrm{d}z}\right)^2 \mathrm{d}z - \frac{1}{2} W_\mathrm{p} \cos^2(\varphi(L) - \varphi_0), \quad (2)$$

where K_{22} is the elasticity constant; W_p is the energy of cohesion with the substrate for $z = L$; and the easy axis at $z = 0$ is directed along the x-axis.

The condition of a minimum of the free energy functional yields the Euler – Lagrange equation and its boundary condition

$$K_{22}\frac{\mathrm{d}\varphi}{\mathrm{d}z} + W_\mathrm{p} \cos[\varphi(L) - \varphi_0] \sin[\varphi(L) - \varphi_0] = 0,$$

$$\frac{\mathrm{d}^2\varphi}{\mathrm{d}z^2} = 0. \quad (3)$$

In addition, because of the infinitely strong cohesion on the rubbed substrate, we have the second boundary condition $\varphi(0) = 0$. The solution of the Euler – Lagrange equation satisfying this condition has the form

$$\varphi(z) = a\,\frac{z}{L}. \quad (4)$$

Here, an unknown constant $a = \varphi(L)$ can be found from the first boundary condition

$$-\frac{2a}{\sin 2(a - \varphi_0)} = \xi_\mathrm{p}, \quad (5)$$

where $\xi_\mathrm{p} = W_\mathrm{p} L / K_{22}$. In the general case, the constant a can be found only numerically.

In the particular case of $\varphi_0 = \pi/2$ (the director tends to be oriented perpendicular to the light polarisation), we have

$$\frac{2a}{\sin 2a} = \xi_2. \quad (6)$$

In this case, a nontrivial solution exists only when $\xi_2 > 1$. This means that for $\xi_2 \leqslant 1$ and the infinitely strong cohesion of the director with the second substrate ($z = 0$), the distribution of the director in the cell will be uniform: $\varphi(z) \equiv 0$. For $\xi_2 \gg 1$, the solution cab be easily found in the form of expansion in $1/\xi_2$:

$$\varphi(z) = \begin{cases} 0, & \xi_2 \leqslant 1, \\ \dfrac{\pi}{2}\left(1 - \dfrac{1}{\xi_2}\right)\dfrac{z}{L}, & \xi_2 \gg 1. \end{cases} \quad (7)$$

Then we obtain the expression for the cohesion energy and its variation during irradiatioin:

$$W_\mathrm{p} = \frac{K_{22}}{L}\,\frac{2\varphi(L)}{\sin 2\varphi(L)}. \quad (8)$$

We studied the cells containing initially the twist-structure characterised by an angle φ', which changed after irradiation. The dashed curves in Fig. 3 illustrate the dependence of the cohesion energy density, calculated from expression (8), on the pump energy (Fig. 3a) and its variation in time after excitation (Fig. 3b). The cohesion energy density corresponding to the initial twisting angle $\varphi' = 18\,^\circ$ is equal to 1.1×10^{-7} mJ cm^{-2}.

Upon irradiation, azo fragments are predominantly aligned perpendicular to the light polarisation. Such an alignment either increases the cohesion energy (the light polarisation is directed along the rubbing direction) or decreases it (polarisation is perpendicular to the rubbing direction). Correspondingly, the parameter a, together with $\varphi(L) = a$, either increases or decreases or even vanishes. Physically, this results in the increase (decrease) or complete vanishing of the director twisting. Thus, one can see from Fig. 3a that upon excitation by a single pulse with the energy density of 80 mJ cm^{-2}, when the polarisation direction coincided with the direction $\boldsymbol{n}$ of rubbing, the director turned by the angle $23\,^\circ$ with respect to the initial orientation angle. The cohesion energy increased by 1.5 times, beginning from the threshold pump energy equal to 30 mJ cm^{-2}. A train of pulses with a repetition rate of 1 Hz formed the static recording for which the cohesion energy density was 2.4×10^{-7} mJ cm^{-2}.

Irradiation by sufficiently intense light causes not only the orientation of azo fragments but also heats the orientant and LC near the surface. This in turn results in a decrease in the cohesion energy due to a decrease in the order parameter both of azo fragments and polymer chains of the film and the LC near the surface. As a result, the orientation ordering of azo fragments are sufficiently high in the central region of the action of a Gaussian beam, the condition $\xi_\mathrm{p} \geqslant 1$ is satisfied, and the rotation angle of the director increases. At the beam periphery, $\xi_\mathrm{p} < 1$ and twisting is absent at all. This is clearly seen upon static recording of a spot by a train of pulses (Fig. 4c).

On the other hand, upon prolonged irradiation by a train of pulses the polymer heating results in its softening, so that the orientation ordering of azo fragments causes the reorientation of the main chain of the polymer. Because the mobility of the main polymer chain at room temperature is rather low, we obtain a new stationary cohesion energy and, correspondingly, a static recording. The characteristic reorientation times of the main chain are rather large, so that the recording contrast can be increased after irradiation ceasing. This explains the time lag of the recording observed in studies of dynamic orientation gratings, which is manifested in the enhancement of the brightness of diffraction maxima within 1 s after irradiation.

4. Conclusions

We have performed the data recording and demonstrated the possibilities of controlling the orientation of the director of a LC cell by nanosecond laser pulses interacting with photosensitive azo fragments of a polymer film deposited onto one of the substrates in the cell. Note the following properties of the reorientation.

(1) The reorientation is characterised by the azimuthal turn of the LC layer director. The azo dye-modified surface can twist LC molecules near the surface by an angle φ'. Irradiation by laser pulses results in twisting or untwisting of the predominant orientation axis depending on the direction of the polarisation vector of radiation and the interaction forces in the volume and surface.

(2) We showed by the method of dynamic holography that this effect develops for ~ 30 μs and the diffraction efficiency of orientation gratings is rather high ($\sim 15\,\%$).

(3) The interference field of laser radiation records in LC cells with azo polymer coating the static gratings with the

spatial frequency no less than 100 lines mm^{-1} at the radiation energy density of 80 mJ cm^{-2}.

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