HYSTERESIS PHENOMENA AND THE EFFECT OF REORIENTATION IN A POLYMER-LIQUID CRYSTAL SYSTEM UNDER THE INFLUENCE OF LASER RADIATION AND UNIAXIAL DEFORMATION

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Abstract. The orientational structure in a polymer-liquid crystal thin film induced by uniaxial mechanical deformation has been studied. It is shown that the dependence of the deflection angle of the director of a nematic liquid crystal on the direction of the tensile force has a hysteresis region in which the orientation structure spontaneously changes from a state with positive torsion of the director to a state with negative torsion. It is established that the effect of modification of the hysteresis loop leads to an expansion of the hysteresis region and a decrease in the field at which orientation bistability appears.

1. Theory

Recently, the search and research of new types of information display devices based on liquid crystals (LC) has been rapidly developing. An important role for the functioning of such devices is played by the phenomenon of reorientation of the predominant direction of molecules (director) in the cell under the influence of external fields (mechanical, electric, magnetic, light, acoustic) [1-4]. The reorientation of the director under the influence of external fields is accompanied by a change in a number of physical parameters that are caused by the absorption or transmission of light radiation [5]. The possibilities of using LC in photonics and optoelectronics become much wider if semiconductor injection lasers based on heterostructures are used as light radiation. To control photon flows, the best option is considered to be the use of polymer-liquid crystal composites (PJCC) [6]. These are thin transparent films containing microscopic capsules of a nematic liquid crystal in volume. This work is devoted to the solution of just such a question.

A nematic liquid crystal (NLC) is a uniaxial liquid. The macroscopic state of the NLC is described by specifying at each point a unit vector n, called the director. In the equilibrium state, the NLC is homogeneous, while inhomogeneous distributions of the director arise in the case of various elastic deformations of the LC. The introduction of the NLC capsule into the composition of the binder (polymer matrix) is carried out by dispersion in various ways (polymerization, cooling of the solution, evaporation of the solvent). These two components are incompatible, so external factors are considered separately. Observation of the structure of the NLC capsule with a polarization microscope makes it possible to obtain bright images occurring under the action of external fields (Fig. 1).
As a rule, the configuration of the director is determined by solving the system of Euler-Lagrange equations [7]. However, if there is a phase transition of the first kind in the system under study, difficulties arise when solving the Euler-Lagrange equations. Therefore, instead of a numerical solution of this equation, an approach based on minimizing the free energy functional is usually used. In this paper, we have focused on the latter option.

For the production of composite films, we used a nematic liquid crystal (NLC) 4-n-heptyl-4’-cyanobiphenyl (7CB) having the following sequence of phase transitions: Cr (-70°C) - LC (28.5°C) – I (42°C). Refractive indices of the studied 7CB: n‖‖=ne,max=1.725 and n┴=n0=1.534 at a temperature T=25°C and a wavelength λ= 0.633 micrometer. The polymer binder was polyvinyl butyral (PVB) with nρ=1.492 [8]. PVB is a soluble polymer characterized by resistance to organic solvents and heat resistance, low gas permeability, good adhesive properties, it provides tangential adhesion to molecules of alkyl cyanobiphenyls derivatives. NLC 7CB was dispersed in a polymer matrix by emulsion technology [9] during evaporation of a common solvent – ethyl alcohol. The prepared mixture was applied to the surface of the glass substrate and the solvent was evaporated in the temperature control mode. The thickness of the samples after drying was 25-30 micrometer, and the capsule size was 7CB 10-15 micrometer. The PJCC films were positioned perpendicular to the direction of the laser radiation passing through the polarizer (Fig. 2) [6].

An experimental setup was used, the peculiarity of which is that an injection laser based on stressed heterostructures is used as a photon source [12]. This class of lasers has advantages related to miniaturization, speed, a wide range of lasers with different wavelength ranges, the ability to work in a wide range of capacities and durability. Of particular importance is the durability of injection lasers, which last up to a million or more hours in continuous operation. By selecting the appropriate heterostructures emitting different wavelengths of radiation, it is possible to investigate the dependence of nonlinear optical processes in these composites on the power and wavelength of laser radiation.

Figure 2 shows a block diagram of an experimental setup that allows us to study the nonlinear optical properties of the PJCC. The device contains a semiconductor injection laser based on InGaAsP/AlGaAs heterostructures operating in the red spectral range, an optical laser beam collimator for producing parallel laser beams, a polymer-liquid crystal composite based on a nematic liquid crystal (NLC) 4-n-heptyl-4’-cyanobiphenyl (7CB), a photodetector for recording the intensity of the laser beam. The entire experimental setup is placed in a special box, which allows you to adjust the ambient temperature.
Figure 2. The scheme of an experimental setup for the study of optical properties of composites based on PJCC. 1-semiconductor injection laser based on InGaAsP/AlGaAs heterostructures, 2-optical laser beam collimator, 3-polymer-liquid crystal composite based on nematic liquid crystal (NLC) 4-n-heptyl-4’-cyanobiphenyl (7CB), 4,5-analyzer, 6-photodetector, 7-computer, 8-adjustable injection laser power supply, 9-thermostat

2. Results and their discussion
Figure 3 shows the dependence of the transmitted optical power of a semiconductor injection laser based on InGaAsP/AlGaAs heterostructures operating in the red spectral range on the magnitude of the relative deformation of the PJCC based on NLC 7CB for the orthogonal component of polarization in the forward and reverse direction of deformation. It can be seen that the phenomenon of optical hysteresis of the power of the transmitted laser radiation is observed at uniaxial deformation. It should be noted that this phenomenon was discovered for the first time for composites based on nematic liquid crystals. Starting from a certain value of the uniaxial stretching value, the dependence of the intensity of the transmitted laser beam will be constant and, starting from this point, there is a change in the direction of uniaxial deformation. As can be seen from Fig. 3, the curves in the forward and reverse directions do not coincide, a hysteresis loop of the laser radiation intensity is observed. The observed effect is associated with relaxation processes of orientation of NLC molecules. Note that when an electric field is applied to the nematic films, the phenomenon of a hysteresis loop of laser radiation intensity is not observed.

It can be seen from Fig. 3 that at a fixed value of T with an increase in Δl/l0, a sharp increase in the rotation angle of the director first occurs, corresponding to an increase in the orientation distortions of the director inside the capsule. However, during the reverse process (i.e. removal of the tensile load), the nature of the curves changes qualitatively. The behavior of the rotation angle of the director inside the NLC 7CB capsule when removing the tensile load near π/2 is described by the formula:

As can be seen from the formula, when removing the tensile load, the angular coefficient dφ/dl is negative, and the angle φ0 is an unambiguous function of Δl/l0. When stretched, the angular coefficient becomes positive and a symmetrical hysteresis loop appears on the curve T=f(Δl/l0), corresponding to the orientation bistability.

With an increase in the angle of rotation of the director, an equilibrium orientation transition, accompanied by a jump in the orientation of the director from a state with positive torsion (Fig. 3, the upper branch of the curve) to a state with negative torsion (Fig. 3, the lower branch), occurs at φ = π/2. The transition of the system, in this case, to a state with opposite torsion, in our opinion, is explained by the fact that the direction of the director in both cases is physically equivalent. In this case, the boundary conditions are symmetric, therefore, the free energies of states with the orientation of the mechanical field φ0=π/2 for positive (φ0>0) and negative (φ0<0) torsion of the director are the same.
The increase in the director's rotation angle corresponding to the behavior along the upper metastable curve in Fig. 3, i.e., an increase in the director's orientation angle in the center of the capsule, leads to an increase in elastic energy. The behavior along the lower curve in Fig. 3, when the condition \( \phi_0 > \pi/2 \) is met, leads to a decrease in elastic energy, since the angle \( \phi_0 \) in absolute magnitude becomes smaller, since the angle between the director and the direction of the mechanical field decreases.

Figure 3. Dependence of the transmitted optical power of a semiconductor injection laser based on InGaAsP/AlGaAs heterostructures on the magnitude of the relative deformation of the PJCC based on NLC 7CB for the orthogonal component of polarization in the forward (\( \bigcirc \)) and reverse (\( \bullet \)) direction of deformation

Further experimental results show that the power of the transmitted laser radiation at a fixed value of the magnitude of the inverse relative deformation gradually increases.

Figure 4 shows the results of measuring the temperature dependence of the growth kinetics of the transmitted optical power of a semiconductor injection laser based on InGaAsP/AlGaAs heterostructures on the delay time at point A in Fig. 3.

The results of calculations of the experimental data obtained show that in the absence of an external field (external tensile force), the deflection angles of the director are zero, which corresponds to the original undisturbed structure. With an increase in the tensile load, the angle of rotation of the director \( \phi_0 \) in the center of the capsule and the angle of rotation of the director at the interface \( \psi_0 \) increase and asymptotically tend to the direction of the external field. These data are more clearly shown in Fig. 4. It can be seen that the dependence of the rotation angle of the director \( \phi_0 \) in the center of the capsule reaches saturation at lower field values than the dependence \( \phi_0 = f(\Delta l/l_0) \); at the same time, the lower the coupling energy \( \sigma \), the greater the difference between the curves for \( \phi_0 \) and \( \psi_0 \), since the polymer matrix is more malleable to the action of the mechanical field than the LC system. In addition, the intensity of III becomes equal to what it had when the condition \( T < T_i \) is met, at the same time decreases to the value of III at \( T < T_i \). This means that the plane of inclination of the NLC 7CB molecules inside the droplet is oriented along the direction of stretching.
Figure 4. Temperature dependence of the growth kinetics of the transmitted optical power of a semiconductor injection laser based on InGaAsP/AlGaAs heterostructures on the delay time at point A in Fig.3.

305K (1); 309K (2); 313K (3)

3. Interpretation of scientific results

Spontaneous polarization of $P_0$ in the nematic layer is perpendicular to the plane of inclination of long molecules [3]. The reorientation of the molecules causes the rotation of the plane of inclination of the molecules and, accordingly, the rotation of spontaneous polarization in neighboring layers. If the thickness of the PJCC sample in the direction of the normal to the composite plane is less than the period of the domains of the crystal lattice, or a mechanical field sufficient to spin the axis of the molecule is applied to the film, the NLC 7CB has a macroscopic spontaneous polarization of $P_0$. In a mechanical field, the polar axis of the NLC 7CB should be oriented along the field (i.e. the axis of strain of stretching), and the plane of inclination of the molecules in the near-surface layers should be perpendicular to the direction of stretching. This is due to the high sensitivity of the NLC 7CB to external influences indicated in the introduction of this work. It should be noted that the effect we found is qualitatively different from the one previously known in NLC 7CB with a reorientation of 180° with respect to the direction of spontaneous polarization [10,11]. This reorientation is not specific to thin films and is observed both in films and in bulk composites, with the plane of inclination of the molecules remaining perpendicular to the directions of spontaneous polarization and the stretching field. In our case, there is a reversal of the structure by 180° with respect to the direction of the mechanical field.
Figure 5. Temperature dependence of the characteristic time of reverse relaxation of the molecules of nematic PJCC, (--) – the limit of the isotropy of NLC

Changes in the characteristic time of the reverse relaxation of NLC molecules of 7CB with an increase in temperature (Fig. 5) characterizes the temperature change in the angle of inclination of the molecules in the capsule volume. The surface ordering is most strongly manifested above the temperature of the phase transition from the crystalline to the nematic state. In this temperature range, the angle of inclination significantly depends on the distance of the domain layer to the capsule surface. In our opinion, a change in the angle of disorientation of molecules in the domains of the 7CB film can cause spontaneous polarization, detected according to Fig. 3.

4. Conclusion
Our comparative analysis showed that flexoelectric polarization may occur in a plane perpendicular to P0, noticeably exceeding the spontaneous polarization of P0 in magnitude, which is associated with a significantly large bending of the director in the 7CB film. With the S-shaped bending of the director, the polarization in the upper and lower parts of the film are directed in different directions. For thin films, the angle of inclination of the NLC 7CB molecules in the center of the capsule may become sufficiently small or higher than the phase transition temperature. In this case, the upper and lower parts of the capsule inside the polymer matrix can be oriented independently, and it will be possible to switch to the S-shaped orientation of the director. The effects we have discovered can be used to record and store information as static memory or bistable state switches.
Low-temperature plasma can be used for the synthesis of various nanostructures and is well suited for the modification of various surfaces. This is shown in many works [13-32].
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