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Physics

EXPERIMENTAL INVESTIGATION OF QUENCHING OF DEFECTS BY EXTERNAL ELECTRICAL FIELD AND ITS INFLUENCES ON PHOTONIC BAND GAP IN CHOLESTERIC LIQUID CRYSTALS

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The possibility of forced quenching of defects in cholesteric liquid crystals with negative dielectric anisotropy has been experimentally examined. It was experimentally shown for the first time that under homeotropic boundary conditions the sharpness of edges of photonic band gap in cholesteric liquid crystals with negative dielectric anisotropy increases in the course of quenching of defects.

Keywords**:** cholesteric liquid crystal, disclination, defect.

Introduction. The creation and investigation of low-threshold, thin, compact and fine devices of modern photonics (linear and nonlinear optics, laser physics etc.) is vital for controlling the properties of light beams as well as of the polarization, direction of propagation, spectral and other properties of laser emissions. Of great interest in this respect are the photonic crystals (PCs) that recently attract more and more growing attention [1], especially those the parameters and properties of which are easily controlled by external influences. With their help the emission and reflection spectra and the polarization of light, passing through the crystal, may be controlled [2]. The above aims are attained by using cholesteric liquid crystals (CLC) that are easily controlled by external fields [3, 4] and resistance to optical emission of which in case of sufficient purity is comparable to that of glass.

Thin films of CLC are similar to nematic liquid crystals (NLC) in planes parallel to their bounding walls (monomolecular layers), i.e., in this plane the molecules are oriented parallel to the substrates along the same axis called the director [5]. At passage from one monomolecular layer to another the director axis of CLC twists by certain small angle α (0.05–0.5^0). Thus it forms a helicoidal structure with the period known as the pitch (P) that is the distance over which a full rotation of 360^0 is completed.

If the periodicity of CLC or, in other words, the pitch of CLC is in visible region of electromagnetic waves, the periodic helicodal structure satisfies the

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conditions for Bragg diffractive reflection of the respective circular polarized light (circular dichroism). The wavelength of Bragg reflection is defined by equation $\lambda_0 = \bar{n} \times p$, where $\bar{n}$ is the average refractive index of CLC. The diffractive reflection from CLC layer takes place only for spectral region $\Delta\lambda = \Delta n \times p$, where Δn is the birefringence of CLC. Only the light (electromagnetic wave) circularly polarized in the direction opposite to the helix is selectively reflected. The selective reflection of light leads to the formation of photonic band gap (PBG) in the transmission spectrum of the light due to, which we can consider CLCs as a one dimensional PC [4].

As was mentioned above, if thin layers of CLC are pure enough, they possess high degree of resistance to optical emission in visible region. However, undesirable scatterings that usually occur due to nonuniformities (orientational defects) in CLC layers, significantly reduce the sharpness of PBG boundaries and, thus, they represent an obstacle for liquid crystal based thin filmed optical devices such as low-threshold lasers [4], light modulators [6], etc., to enter to the phase of technological applications [7]. Requirements are especially stringent in lasers and modulators (when used particularly in imaging systems), because minor scatterings can significantly reduce the instrument's quality-factor.

The goal of the present work is experimental investigation of the influence of external electrical field on nonuniformities (disclinations and orientational defects) existing in CLCs with negative dielectric anisotropy and the possibility of quenching of those nonuniformities. The influence of the external electric field on PBG has been also investigated.

The nonuniformities of two types were investigated. First ones are the disclinations, which are rupture lines of optical homogeneity of the material. Molecules abruptly change (in space) their orientations within these lines. As in case of nematic liquid crystals (NLC) the molecules conserve the direction of orientation on both the sides of disclinations, the rotation of director within the line can not be random, but must be multiple of π. In NLC cells the disclinations usually arise, when for some reasons (inhomogeneity of the boundary conditions at the substrates, convective fluxes, thermal nonuniformities etc.) the molecules are forced to abruptly change (in space) their preferred sense along which, their long axes (director) are directed [8, 9]. In case of CLCs the disclinations exist almost invariably (Fig. 2, a and b), they exist even in case of the absence of above mentioned structural changes. The disclinations can move, unite or split with time [9–11]. The second type of nonuniformities is connected with bad orientations and they exist in the whole bulk of the cell.

Experiment. The discussed two types of nonuniformities are comparable in size with the wavelength of visible light and thus, they cause undesirable scattering of visible light. In order to observe the feasibility of ‘‘clearing’’ the nonuniformities by application of electrical field, we have used planar and homeotropic cells, filled with CLC having negative dielectric anisotropy [6] ($\Delta\varepsilon = \varepsilon_{\parallel} - \varepsilon_{\perp}$, where $\varepsilon_{\parallel}$ and $\varepsilon_{\perp}$ are dielectric constants of material in directions parallel and perpendicular to the long molecular axis respectively).

It is well known that the long axes of the molecules of liquid crystal (LC) with negative dielectric anisotropy are arranged perpendicular to the applied electric field. This means that instead of destruction of the helicoidal periodic structure, the applied external electrical field stabilizes the structure. As (in the ideal case) the molecules in a helicoidal structure are always parallel to the particularly cell substrates, then molecules with different orientations (those located in the disclination lines) will reorient under the influence of electric field and be included in the helicoidal structure. Moreover, one can expect that the possibility of molecular fluctiations in the direction, perpendicular to the monomolecular layers, will decrease. So, one can suppose that under the influence of applied electric field the deviation inclinations of molecules from the periodic, helicoidal structure (defects) will decrease. Consequently, undesirable diffusion caused by director fluctuations will also decrease.

CB15 (purchased from ‘‘Merck’’ company) CLC with negative dielectric anisotropy, which possesses a PBG in the visible region of light (~ 520–560 *nm*) was used in experiments.

The situation is more clear in case of planar orientation: the molecules in the layers which are in contact with substrates are oriented parallel to substrates and point their axes in certain (rubbing) direction. The molecules in next layers also have a certain preferred direction that is also parallel to substrates, but it slightly twists from layer to layer forming thus a twisted (helicoidal) periodic structure [9] (Fig. 1, a). However, in CLC cells some defects almost always exist in the form of disclinations [9] that usually cause undesired diffusion of the light. Usually, the mentioned disclinations disappear in the course of time, but these time periods may be long enough (days, weeks…). Microscopic images of 5 *μm* thick cell are presented in Fig. 2, when the cell is placed between crossed polarizers. The image in Fig. 2,a is taken immediately after the cell fabrication. One can see, the sufficient disclination lines that gradually disappear during days (Fig. 2,b). The disclinations disappear rapidly, if we apply electric field (perpendicular to glass substrates). Indeed, as is seen in Fig. 2,c, which is taken 5 *s* after the application of field, the molecular structure is rather uniform without any disclination and does not change after switching off the electrical field.

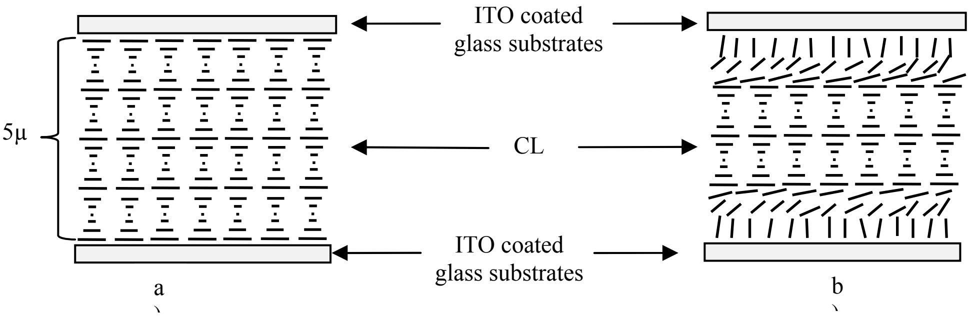

Fig. 1. Schematics of molecular organization in CLC cells: a) planar and b) homeotropic boundary conditions on limiting substrates.

From the point of view of uniform molecular orientation, the situation becomes more complex in CLC cell with homeotropic boundary conditions. The molecules of the near substrate layers, orient their long axes perpendicular to substrates and tend to transfer this direction to the layers in neighborhood. But as it is already mentioned, CLC molecules tend to form a helicoidal structure. Thus, the forces of surfactant layer coated on the substrate and defining orientations of molecules appose intermolecular interactions tending to form a helicoidal structure. As a result, an original arrangement of molecules is formed inside the cell. The molecules in direct contact with glass substrates align perpendicular to them. In the next layers, because of weakening of the influence of the surfactant layer, the director gradually turns up to parallel orientation (to substrates). Finally, a helicoidal periodic structure is formed, as is the case in planar cells (Fig. 1, b).

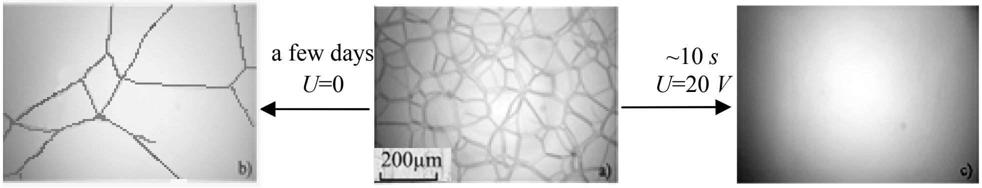

Fig. 2. Images of 5 *μm* thick CLC cell with planar boundary conditions, placed in the field of "Zeiss" polarizing microscope. Images are taken: a) immediately; b) few days after the cell fabrication; c) about 10 *s* after applying $U=20\ V$ voltage.

As is seen from the polarizing microscope images (Fig. 3, a), the nonuniformities in homeotropic cell exceed in number those in planar cell (Fig. 2). Here, along with the disclination lines there are also numerous little nonuniform domains that are dispersed in the whole volume of the cell. According to the experiments, the applied electrical field (20 *V* to 5 *μm* thick cell) quenches the disclinations and significantly reduces the dimensions of nonuniformities (Fig. 3, b). Usually, the disclination lines are not oriented but are dispersed in the CLC cell. In Fig. 3,a the lines have some preferred direction, because of the flow of CLC during the filling of the cell with CLC (the image is taken immediately after the cell fabrication). The structure recovers from Fig. 3,a state to the Fig. 3,b state (but without any preferred direction of disclination lines) in several days after switching off the electrical field.

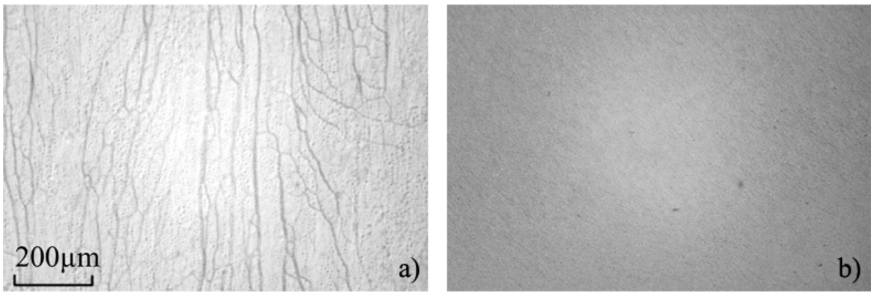

Fig. 3. Images of the 5 *μm* thick CLC cell with homeotropic boundary conditions, placed in the field of "Zeiss" polarizing microscope: a) immediately after making the cell; b) about 10 *s* after applying $U=20\ V$ voltage.

To study the variation of PBG concurrent with the quenching of disclinations (caused by external electric field), we have also realized a spectroscopic investigation of the cell. The transmission spectra of homeotropic and planar cells for different applied voltages are shown in Fig. 4. According to the experiments, the influence of external electric field on the transmission spectrum of the planar CLC cell is not significant despite the fact that the field eliminates the structural nonuniformities. The PBG is always clearly distinguished (Fig. 2, a, b, c).

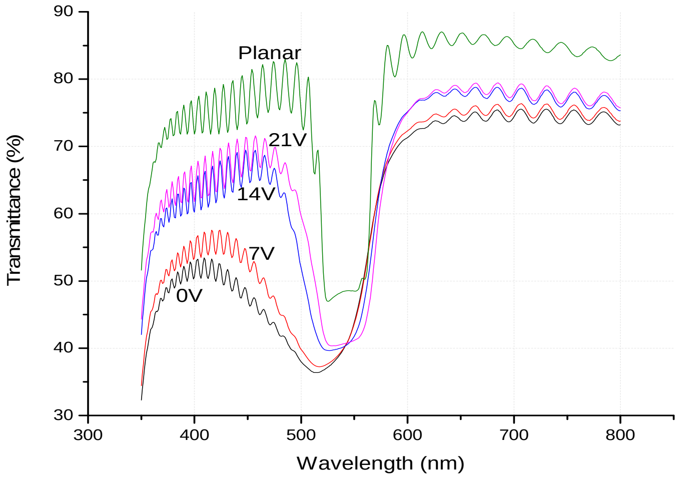

Fig. 4. Spectroscopic observation of band gap of 5 *μm* thick planar and homeotropic CLC cells. Different voltages are applied between homeotropic cell substrates (0; 7; 14; 21 *V*).

The image is different in case of homeotropic cell. As is seen from the Fig. 4, the transmission spectrum of the homeotropic cell void of the voltage (U=0) differes from that of the planar cell. The PBG is warped and the wings are not sharp enough. It is important to mention that compared with the transmission spectrum of an “ideal” helicoidal periodic structur’s (as that of the planar cell) the differences are more clear in the short wavelength wing and in shorter regions. The applied electrical field “cleans” the transmission spectrum, thus bringing it towards the spectrum of ideal helicoidal structure. As is seen from the Fig. 4, the transmission spectrum of the homeotropic cell for U=21 V applied voltage is close enough to that of the planar cell. In that case, one can clearly distinguish the PBG boundaries, which is enough symetric, as in the case of the planar cell.

Discussion. According to the theory of molecular interactions, the forces of the interaction of CLC molecules among themselves and interaction of CLC molecules with the ones on the substrate surface are important for the orientational order of CLC molecules. At the application of electric field to the CLC film, the forces exerted by this field also contribute to the rearrangement of CLC molecules. In the equilibrium state, the contributions of momenta produced by aforementioned three forces are balanced.

The interaction forces acting between CLC molecules tend to form a helicoidal (twisted) periodic structure. The interaction forces between CLC molecules and those on confining layers are usually short-range.

In the planar cell, these two interractions force the molecules to form helicoidal structure, pointing their axes in plains parallel to the substrates. In case of homeotropic boundary conditions on the substrates the molecules that are in direct contact with them orient their long axes perpendicular to substrates, thus making the structure unstable. For this reason, sometimes CLC forms a helicoidal periodic structure, the axis of which is parallel to the substrates confining the cell.

One can conclude thus that in case of positive dielectric anisotropy $(\Delta\varepsilon > 0)$ the applied external field, which is perpendicular to glass substrates, disturbes the periodic structure of CLC and starting from a certain value of electric field the CLC cell passes to the homeotropic state [12, 13]. As was already mentioned above, the situation is different when the cell is filled with negative dielectric anisotropy $(\Delta\varepsilon < 0)$ CLC: under the applied external electric field the axes of CLC molecules arrange perpendicular to the direction of electric field contributing thus to the formation of a helicoidal structure of CLC with axis perpendicular to the substrates (planar helicoidal structure).

The experiments show that even in the cell subject to the planar boundary conditions, in the absence of electric field there arise some nonuniformities (Fig. 2, a, b). This may be due to the nonuniformities of the boundary conditions on the confining substrates, the presence of foreign particles in CLC, appearance of hydrodynamic flows owing to diverse reasons etc. As is seen in the image taken with the polarizing microscope (Fig. 2, c), at the application of electric field with 4 *V*/*μm* intensity the reasons causing the rise of disclinations are eliminated. As a result, a CLC cell with sufficiently uniform distribution of LC molecules is obtained.

In case of homeotropic cell the aforementioned three forces acting on the CLC molecules are at variance from the viewpoint of helicoidal structure formation. The forces exerted by the substrates destabilize the structure of CLC, the helical axes of which are oriented normal to the substrates. As a result, in the absence of electric field one can see in Fig. 3,a some spot defects spread over all the bulk of cell along with the disclination lines. Here also, as in the planar cell, the electric field tends to reorient the molecules that cause the nonuniformities to the helicoidal structure, and as we see in Fig. 3,b, the electric field eliminates the disclinations and reduces the size of spot defects.

For qualitative description of the above phenomenon we can avail of the graphs shown in Fig. 4. It is seen that prior to the application of electric field (U=0) the transmission spectrum of homeotropic cell significantly differed from that of the planar cell, and as was already noted the difference is larger in the short wavelength part of PBG. It can be accounted for the presence inside the cell of small helicoidal formations (domains), the axes of which are not perpendicular to the cell walls. So, inside the cell there exist numerous little helices, the axes of which are not co-directed, but make different angles with the cell wall. As a result, 1 *mm* spectrometer beam impinged against numerous little helices having different orientations, and owing to that the reflection of not only PBG range, but also shorter wavelengths brought to seeming widening of PBG. The application of electric field to CLC layer leads to reorientation of above little helixes (that orient their axes perpendicular to the confining substrates). The deviation from the normal incidence decreases and, hence, the short wavelength part of PBG in the transmission spectrum increases. As is seen in Fig. 4, after application of E=4.2 *V*/*μm* strength electric field (*U*=21 *V*) the wings of spectrum are rather symmetric, which means that there are no (or little) tilted helices in the bulk of cell. At the application of *U*=21 *V* the depth and width of PBG increased by the factor of 1.8 and decreased by the factor of 1.4 respectively. However, experiments show that the transmission spectrum of homeotropic cell is still lower than that of planar cell, which means that the orientations of molecules are still homeotropic near the substrates (because of strong anchoring of molecules to substrates).

Conclusion. So, in the present work it was experimentally shown that the orientational order of CLC is increased by using negative dielectric anisotropy CLC and application of electric field. Under influence of electric field the nonuniformities in the director distribution decrease and the scattering of light in the CLC layer is reduced. It was experimentally shown that under homeotropic boundary conditions the sharpness of the edges of photonic forbidden band in negative dielectric anisotropy CLC cells is increased and its width decreased as the defects are quenched. The results can be used for numerous other investigations as well as in CLC devices, e.g., in CLC based light modulators [6].

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Կ.Ռ. Ալլահվերդյան. Խոլեստերիկ հեղուկ բյուրեղներում արտաքին էլեկտրական դաշտով արատների ճնշման և ֆոտոնային արգելված գոտու վրա դրա ազդեցության փորձարարական հետազոտությունը

Փորձնականորեն հետազոտվել է բացասական դիէլեկտրական թափանցելիության անիզոտրոպիայով խոլեստերիկ հեղուկ բյուրեղներում արատների արտաքին էլեկտրական դաշտով ստիպողական ճնշման հնարավորությունը: Առաջին անգամ փորձնականորեն ցույց է տրվել, որ հոմեոտրոպ սահմանային պայմաններով խոլեստերիկ հեղուկ բյուրեղներում արատների ճնշմանը զուգընթաց աճում է ֆոտոնային արգելված գոտու սահմանների կտրուկությունը:

***К.Р. Аллахвердян*. Экспериментальное исследование подавления внешним электрическим полем дефектов и его воздействия на фотонно-запрещенную зону в холестерических жидких кристаллах**

Экспериментально исследовано вынужденное внешним электрическим полем подавление дефектов в холестерических жидких кристаллах с отрицательной анизотропией диэлектрической проницаемости. Впервые экспериментально показано, что в холестерических жидких кристаллах с гомеотропными граничными условиями в ходе подавления дефектов увеличивается резкость границ фотонно-запрещенной зоны.