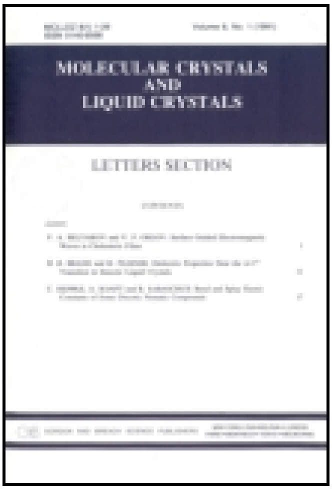

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Manifestation of the Chiral Liquid Crystal Boundary Conditions in Lasing Features

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Manifestation of the Chiral Liquid Crystal Boundary Conditions in Lasing Features

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Spectral, thresholds and spatial characteristics of lasing in dye-doped chiral liquid crystals (CLC) of steroidal - type for parallel and perpendicular orientation of directors on the substrates are studied. The observed improvements of lasing parameters can be supposedly explained by appearance of the helical structure defect. Corresponding details of theoretical analysis in the frame of the similar model is presented. The observed lasing behavior of CLC with orthogonal director orientation occurs to be similar to predicted ones in the theory for the oscillation on the defect mode in the presence of phase defect layer in the helical structure.

Keywords Cholesteric liquid crystal; distributed feedback laser; photon crystal; defect mode

1. Introduction

Dye-doped CLC with natural spiral layer ordering structure present of great interest for various lasing devices. Combining CLC and organic dyes in the same matrix allows fabrication of microlasers with distributed feedback (DFB) which oscillates due to Bragg scattering and amplification of the dye emission on helical periodic structure [1–2]. This feature makes similar DFB-lasers promising element for performance a display of highest brightness.

It was discussed in [3] that different theoretical models depending on matrix birefringence might explain the lasing features in CLC. For the steroidal CLC made of cholesterol ether mixture with the small birefringence ($\Delta n \approx 0{,}04$) spectral and energy characteristics of lasing can be explained in frame of the coupled wave model [4–5]. This model predicts the lasing spectrum position at the SR band center, the strong selection of longitudinal modes and low energy output in the case of the strong wave coupling. Recent studies of lasing thresholds and lasing spectra in steroidal CLC as function of the planar texture perfection have confirmed that lasing in these systems can be explained by the coupled wave model [3].

At the same time for the induced CLC created by means of twisting dopants in a nematic liquid crystal (NLC) ($\Delta n \approx 0{,}1 \div 0{,}25$) lasing characteristics show some peculiar features

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that requires other model for description. That are the location of the lasing spectrum at the edge of selective reflection (SR) band, higher energy output, lower selectivity (oscillation on more than three longitudinal modes [6], which was never observed in steroid CLC [1–3]), that cannot be described by the coupled waves model. These features. were explained by the lasing model on the base of photonic crystal [7 -8] cavity with specifically edge modes [9]. The similar lasing model for induced CLC was proposed firstly in [6] and developed in [10–11]. Results of experimental studies of lasing in induced CLC are presented in several review papers [15,16].

However, the influence of defects on the lasing characteristics in the steroidal CLC, that was theoretically considered in [10–11], experimentally was not studied. The possibility and conditions of lasing on defect mode in such CLC presents the main subject of interest. In this work we contribute the experimental results concerning the spectral, thresholds and spatial characteristics of lasing in dye-doped CLC of a steroidal-type for two cases of mutual orientation of their directors on the surface of the orienting substrates (parallel and perpendicular). We presents also the results of a theoretical study of the defect modes in such texture and its manifestation in the lasing spectral and spatial characteristics.

2. Samples and Experimental Set-Up

As the matrix of the steroidal CLC 3-component mixture containing 42,5% Tekon-20 (analogue of cholesteryl oleate), 35% cholesteryl pelargonate, and 22,5% cholesteryl chloride (produced in Institute of Single Crystals NAS of Ukraine) with temperature alteration of spiral pitch $\sim$3 nm/$^\circ$ C was used. CLC was doped with the phenolenone dye F490 (NIOPIK, Moscow, Russia) with weight concentration 0,2–0,3%. The planar texture was created by means of the standard technique [3]. The technology includes rubbing of the glass or quarts substrates coated with a layer of SnO_2 and polyimide lacquer (PAK) and their further mutual shifting in the rubbing direction at the phase transition temperature (after filling of the CLC plane capillary). In the manufacture of textures with orthogonal director orientations on substrates instead of their shift after sample cooling, we used a small reversal of one of the substrates at an angle of $\approx$20° and back.

The lasing parameters were investigated by means of the experimental setup described in [3]. Optical pumping of the doped CLC was carried out by the second harmonic ($\lambda =$ 530 nm) of a Q-switched Nd $^{3+}$ laser operating in a slow pulse repetition mode with the pulse duration $\approx$20 ns. The second harmonic beam focused by a lens with focal distance of 21 cm on the CLC- sample in a spot with a diameter $\approx$1,0 mm. A maximal power density of the second harmonic radiation was $\approx$30 MW/cm^2 and attenuated by neutral filters. The lasing spectra of the dyed CLC corresponding to each pumping pulse were optically imaged in a focal plane of a spectrograph with an inverse dispersion 0.6 nm/mm and then displayed by the video camera on a PC monitor.

Transmission spectra of the CLC were measured at room temperature by spectrophotometer SF-20 (LOMO, St. Petersburg). The fluorescence spectra of dye in CLC were measured by spectrometer MPF-4 “Hitachi”.

3. Results and Discussion

Figure 1 shows the spectral characteristics of the planar texture CLC samples based on above-mentioned ternary mixture of cholesterol esters in the parallel orientation of directors

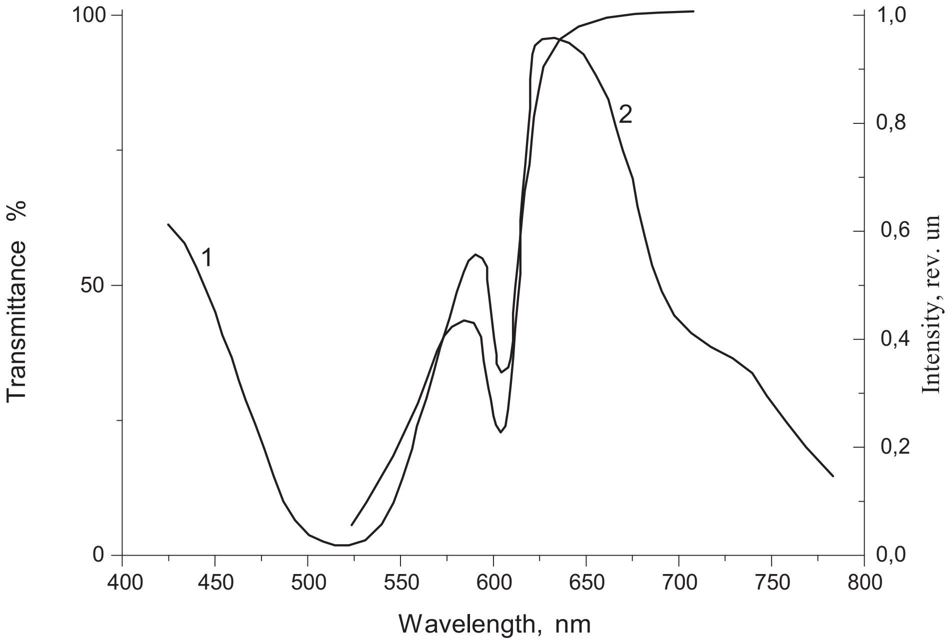

Figure 1. Transmission spectra for linearly polarized light (1) and fluorescence spectra for diffracting left circular polarization (2) of dye-doped CLC at parallel director orientation on the substrates. The layer thickness is 45 μm.

on the substrates. Observed minima in the transmission spectrum (curve 1) are due to absorption of the dye (525 nm) and selective reflection (SR) band of the CLC (600 nm).

The fluorescence spectrum of the impurity CLC (curve 2) was recorded along the helix axis using an achromatic quarter-wave plate and a polarizer to select the diffracting left - hand circular polarization. The dip in the fluorescence spectrum, which was described firstly in [1], coincides with the SR spectrum of the CLC and formed as result of the suppression fluorescence in this frequency range due to the photon localization in the periodic structure of the CLC. For the planar texture of CLC formed by the perpendicular orientation of directors at orienting substrates, the transmission and fluorescence spectra are the same as shown in Fig. 1., except that the half-width of the SR spectrum at a crossed orientation of directors is reduced by 7%.

For relatively high-quality textures with parallel director orientation on the substrates, lasing spectra of steroidal CLC is characterized by the presence of three longitudinal modes, the power of which increases after threshold linearly over the entire range of excitation up to the degradation of the planar texture due to the temperature increase in active zone and the corresponding phase transition in the liquid state [17] (Fig. 2a,). Note that for this orientation of directors we recorded the lowest lasing threshold.

The spatial pattern of radiation for the considered case looks like following: along with an intense central core, there are modes of the ring structure, which increases the total beam divergence up to 18 degrees at layer thickness of 45 μm (Fig. 2b,). The most bright ring structure is observed at a layer thickness L = 40–50 μm, but at a layer thickness L > 60 μm as well as in wedge like samples with angle $\approx 1^\circ$ the ring structure is blurred. It was observed that the number of the rings and the angle divergence are rapidly increases with the

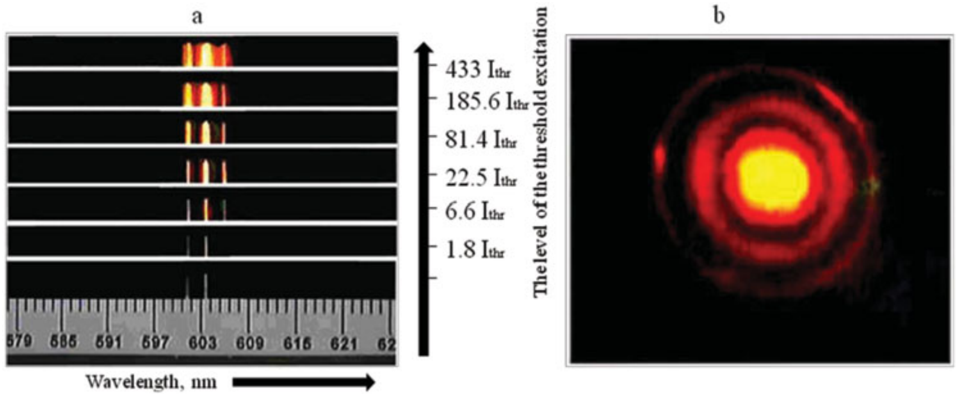

Figure 2. Spectral characteristics of lasing in the steroidal CLC with parallel director orientation on the substrates. by varying the intensity of the excitation (a) and the spatial pattern of lasing (b) by the excitation intensity at 50 $I_{thr.}$. The layer thickness is 45 μm.

pumping growth up to Ip $\approx$ 10 Ithr and are not changed with the further pumping growth. The identity of the lasing spectral composition in the rings and the central core, registered in [18], can be considered responsible for its formation due to the core beam scattering by optical inhomogeneities of the active medium. This scattering after multiple reflection on the cavity mirrors, accompanied by the formation of the lasing modes propagating at discrete angles to the axis of the resonator [19].

This explanation of the ring structure origin has recently received confirmation in the experiments [20], which as a result of precision measurements on an improved apparatus has been found that there are small differences in the frequencies belonging to different rings, that is characteristic of transversal modes.

Thus, in a modern interpretation the ring structure in the spatial pattern of the DFB laser emission on CLC is due to the generation of transverse circular modes of Laguerre -Gaussian type.

For textures with perpendicular orientation of the directors on fused quartz substrates in the spatial pattern of lasing is observed no rings, only the central core (fundamental transverse mode), the spatial divergence of which being about 3 angular degrees. In the lasing spectrum in the case it was registered only the lowest threshold longitudinal mode, the spectral width is reduced by more than an order of magnitude. The relationship of the spectral and spatial characteristics of lasing in a steroidal CLC appears in the followi ng way: as soon as the lasing spectrum reveals higher longitudinal modes, in the spatial pattern of lasing arises a ring structure. In spite of the fact that reflection dip in the SR bands as sign of the defect mode [10–11] was not found, but the spatial characteristics of lasing, namely the absence of a ring structure typical, makes it possible to suggest the presence of the defect structure under the cross orientation of its directors. The phase jump in this texture defined by the substrates can be appeared through the thickness of the layer, thus causing a defect mode.

4. Theory

In the calculation was used the results of [10, 11] for the case of the active layer and an isotropic layer and nonabsorbing CLC layers with the introduction of appropriate changes

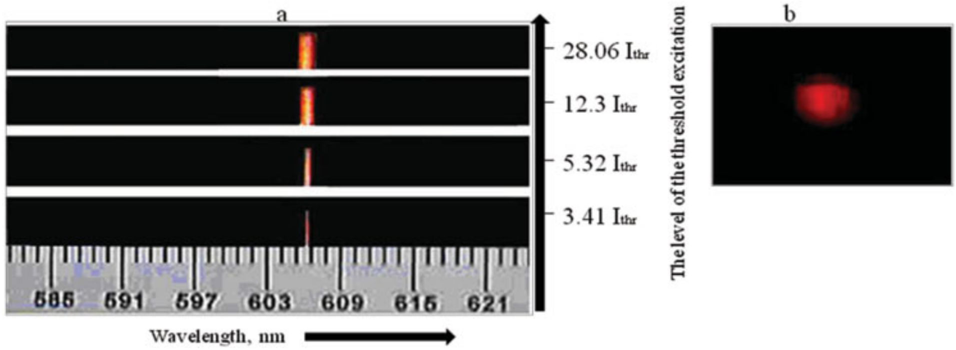

Figure 3. Spectral characteristics of lasing in the steroidal CLC with orthogonal director orientation under varying pumping (a) and the spatial pattern of lasing (b) by the excitation intensity at 28 $I_{thr.}$. The layer thickness 45 μm.

in the formulas obtained in [10, 11]. The assumptions of [10, 11] that the average dielectric constant of CLC ε_0 is coinciding with the dielectric constant of a defect layer and external media so upon reflection at the boundaries the polarization conversion is absent.

The transmission $|T(d, L)|^2$ and reflection $|R(d, L)|^2$ intensity coefficients (of light of diffracting circular polarization) for the whole structure may be presented in the following form:

$$|T(d, L)|^2 = |[T_e T_d \exp(ikd(1 + ig))]/[1 - \exp(2ikd(1 + ig))R_d R_u]|^2 \quad (1)$$

$$|R(d, L)|^2 = |\{R_e + R_u T_e T_u \exp(2ikd(1 + ig))/[1 - \exp(2ikd(1 + ig))R_d R_u]\}|^2 \quad (2)$$

where Re(Te), Ru(Tu) and Rd(Td) are the amplitude reflection (transmission) coefficients of the CLC individual layer for the light incidence at the outer (top) layer surface, for the light incidence at the inner top CLC layer surface from the inserted defect layer and for the light incidence at the inner bottom CLC layer surface from the inserted defect layer, respectively. It is assumed in the deriving of Eqs. (1, 2) that the external beam is incident at the structure from the above only. The factor (1 + ig) is related to the defect layer only and corresponds to the dielectric constant of the defect layer having the form ε_0 (1 + 2ig) with a small g being positive for an absorbing defect layer and negative for an amplifying one.

The defect mode frequency ω_D is determined by the following dispersion equation:

$$\{\exp(2ikd(1 + ig)) \sin^2 qL - \exp(-i\tau L)[(q\tau/\kappa^2)\cos qL + i\left((\tau/2\kappa)^2 + (q/\kappa)^2 - 1\right)\sin qL]^2/\delta^2]\} = 0 \quad (3)$$

where $\kappa = \omega_0{}^{1/2}/c$, $\tau = 4\pi/p$, p is the cholesteric pitch, $\delta = (\varepsilon\parallel - \varepsilon\perp)/(\varepsilon\parallel + \varepsilon\perp)$ is the dielectric anisotropy, and $\varepsilon\parallel$, $\varepsilon\perp$ are the local principal values of the LC dielectric tensor, d is the thickness of defect layer,

$$q = \kappa\{1 + (\tau/2\kappa)^2 - [(\tau/\kappa)^2 + \delta^2]^{1/2}\}^{1/2}.$$

For finite thicknesses of CLC layers L ωD occurs to be a complex quantity which may be found by a numerical solution of Eq. (3).

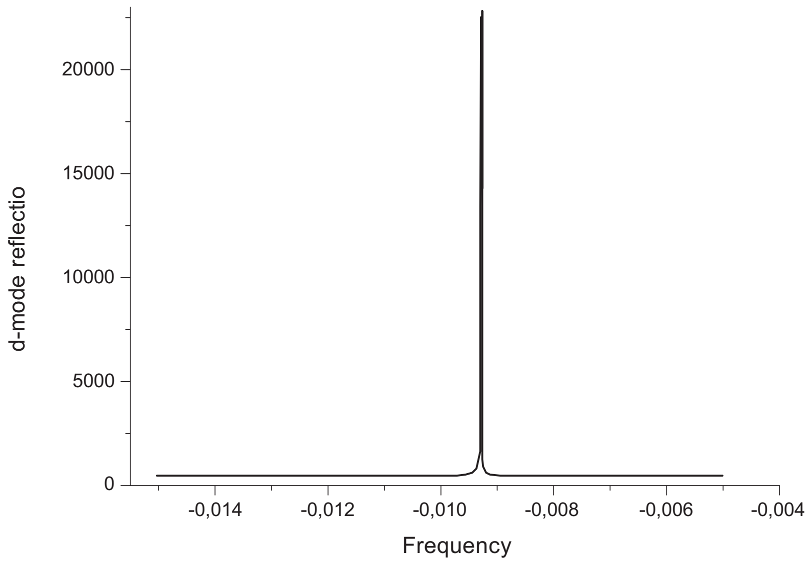

Figure 4. R(d) versus the frequency for amplifying defect layer and nonabsorbing CLC layers at g = −0.04978 for d/p = 0.01; $\delta = 0.05$, N = 33.

The analytic approach for thick CLC layers ($|q|L > 1$) results that for ω_D in the middle of the stop-band the threshold gain is given by the expression:

$$g_t = -(2/3\pi)(p/d)\ \exp[-2\pi\delta(L/p)] \tag{4}$$

So, as the formula (4) shows the thinner defect layer is the higher is threshold gain g.

The dispersion equation, describing the defect mode with an active defect layer has been analytically and numerically solved. The results show that the minimum threshold lasing corresponds to the location of the defect mode frequency in the middle of the SR band and lasing threshold is inversely proportional to the thickness of the defect layer. Thus, the optimum for the experimental study is a situation in which a planar defect is in the middle of the CLC layer.

5. Conclusions

In spite of that dips in SR bands as sign of the defect mode [10, 11] in the texture under study was not found, the spatial characteristics of lasing, namely the absence of a ring structure ty pical for the CLC with a defect mode, makes it possible to suggest the presence of the defect structure in the cross orientation of its director at the layer boundaries. The phase jump in this texture defined by the substrates can be moved to the inner part of the layer, thus causing a defect mode.

The dispersion equation, describing the defect mode with an active layer of a defect has been analytically and numerically solved. The results show that the minimum threshold lasing corresponds to the location frequency of the defect mode in the middle of the SR band and lasing threshold is inversely proportional to the thickness of the defect layer.

In conclusion we recognize that presented explanation is preliminary and more plain CLC structures with defects it is necessary to create in order to realize lasing system with the best output lasing parameters.

References

[1] Ilchishin, I. P., Tikhonov, E. A., Tishchenko, V. G., & Shpak, M. T. (1980). *JETP Lett.*, *32*, 24.
[2] Ilchishin, I. P., Kleopov, A. G., Tikhonov, E. A., & Shpak, M. T. (1981). *Bull. Acad. Sci. USSR. Phys. Ser.*, *45*, 13.
[3] Ilchishin, I., Tikhonov, Eu., & Belyakov, V. (2011). *Mol. Cryst. Liq. Cryst.*, *544*, 178.
[4] Kogelnik, H., & Shank, S. V. (1972). *J. Appl. Phys.*, *43*, 2327.
[5] Kneubuhl, F. K. (1983). *Infrared. Phys.*, *23*. 115.
[6] Kopp, V. I., Fan, B., Vthana, H. K. M., & Genack, A. Z. (1998). *Opt. Lett.*, *23*, 1707.
[7] Yablonovitch, E. (1987). *Phys. Rev. Lett.*, *58*, 2059.
[8] John, S. (1987). *Phys. Rev. Lett.*, *58*, 2486.
[9] Dowling, J. P., Scalora, M., Bloemer, M. J., & Bowden, C. M. (1994). *J. Appl. Phys.*, *75*, 1896.
[10] Belyakov, V. (2008). *Ferroelectrics.*, *494*, 127.
[11] Belyakov, V. A., & Semenov, S. V. (2011). *JETP*, *112*, 694.
[12] Kopp, V. I., Zhang, Z.-Q., & Genack, A. Z. (2003). *Prog. Quant. Electron.*, *27*(6), 369.
[13] Palffy-Muhoray, P., Gao, W., Moreira, M., Taheri, B., & Munoz, A. (2006). *Phil. Trans. R. Soc.*, A.364, 2747.
[14] Coles, H., & Morris, S. (2010). *Nature Photonics*, *4*, 676.
[15] Takezoe, H. (2012). In: *Liquid Crystals Beyond Displays: Chemistry, Phys ics, and Applications*. Quan Li (Ed.), Chapter 1. John Wiley & Sons, Inc.
[16] Blinov, L. M., & Bartolino, R. Eds. (2010). *Liquid Crystal Microlasers*. Chapter 1. Transworld Research Network, Kerala, India.
[17] Ilchishin, I. P. (1996). *Bulletin Russian Academy Sciences. Phys.*, *60*, 494.
[18] Ilchishin I. P., Tikhonov E. A., & Shpak M. T. (1988). *Ukrainian Phys. Journ.*, *33*, 10 [in Russ].
[19] Stoichev, B. P., & Szabo, A. (1963). *Appl.Opt.*, *2*, 811.
[20] Palto, S. P., Shtykov, N. M., Umansky, B. A., Barnik, M. I., & Blinov, L.M. (2006). *Opto-Electronics Review*, *14*, 323.