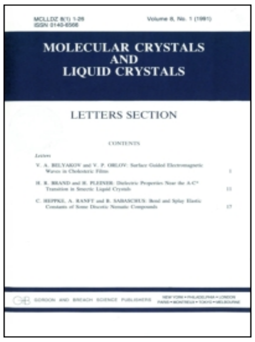

Taylor & Francis
Taylor & Francis Group

Molecular Crystals and Liquid Crystals

ISSN: 1542-1406 (Print) 1563-5287 (Online) Journal homepage: http://www.tandfonline.com/loi/gmcl20

Dye-doped CLC-lasers: Interdependence of spectral and angular characteristics and boundary conditions of the active layer

I. P. Ilchishin, E. A. Tikhonov & T. V. Mykytiuk

MOLECULAR CRYSTALS AND LIQUID CRYSTALS
2016, VOL. 639, 47–54
http://dx.doi.org/10.1080/15421406.2016.1254518

Dye-doped CLC-lasers: Interdependence of spectral and angular characteristics and boundary conditions of the active layer

I. P. Ilchishin, E. A. Tikhonov, and T. V. Mykytiuk

Institute of Physics, National Academy of Sciences of Ukraine, Kyiv, Ukraine

ABSTRACT
We have studied the spectral and angular emission characteristics of a cholesteric liquid crystal (CLC) laser under specific conditions of its planar orientation. It is discovered that the orthogonal orientation of directors on substrates greatly improves the lasing spectrum and the angular divergence. A shift of the lasing spectrum and an increase in the lasing threshold under such conditions can testify to the presence of a defect in such structure, though no dip in the spectrum is registered. The origin of the ring structure of a laser beam is studied, and it is established that this structure is caused by the excitation of the lasing on higher axial modes.

KEYWORDS
cholesteric liquid crystal; planar texture; lasing spectra; angular divergence

1. Introduction

Dye-doped cholesteric liquid crystals (CLC) with natural periodic structure stay promising for investigations [1]. As a result of the development of studies in this direction, various types of similar lasers have been created till now. One of the advantages of a CLC-laser is a possibility to create its active medium of arbitrary form and area, which makes it promising for displays of high brightness. The studies of such lasers and directions of their possible applications took place at many research centers and had led to a number of significant results. The extended spectral range of lasing from the visible to the ultraviolet region had been demonstrated [2]. An extremely low threshold of lasing to be tens of nanojoules had been reached [3]. A CW lasing in a cholesteric with laser pumping had been realized as well [4]. A fast method of frequency phototuning of the lasing (six milliseconds for one nanometer) was developed [5]. Obtained lasing of quantum dots in a cholesteric in the near infrared region [6]. The cause for the dependence of the energy efficiency in such lasers on the intensity of excitation and the order parameter of a doped dye was clarified [4, 7]. In works [8] and [9], it was shown that a similar dependence is related to the induced thermal grating, which is in the opposite phase to the main refractive index grating. These features in the energy efficiency of a CLC-laser, for example, the nonlinear dependence of the the laser energy on the excitation energy, are observed for all types of CLC-lasers and can be explained within the model of coupled waves [10]. The model of photonic crystals, which was first used in [11] to explain the features of the laser spectra of nematics with induced helix, cannot explain features of the energy efficiency of a CLC-laser.

CONTACT T. V. Mykytiuk mykytar@iop.kiev.ua Institute of Physics, National Academy of Sciences, 46, Prosp. Nauky, Kyiv 03680, Ukraine.
Color versions of one or more of the figures in the article can be found online at www.tandfonline.com/gmcl.

So, a number of important results were obtained, which allows one to produce the active media of these lasers at a high-quality level. Among them, the use of polymeric CLCs for the lasing [12, 13] and the introduction of various defects in the helical structure, which allows one to optimize some characteristics of a CLC-laser, should be mentioned [14–15].

However, for these laser types, there exist a number of obstacles to optimize the lasing characteristics. Due to the small thickness of the active layer, the laser radiation is characterized by a large angular divergence of tens of degrees. The small thickness of the active layer leads to a relatively broad spectrum of the multimode lasing. Disputable is the nature of a ring structure in the beam of a CLC-laser. The further detailed study is required for the influence of the quality of a planar texture on the lasing characteristics of such laser.

The aim of the present work is to investigate the connection between the spectral and spatial characteristics of a CLC-laser, the orientation of directors on orienting substrates, and the quality of a planar texture for the further optimization of these characteristics. The objective of the work is also the clarification of the nature of a ring structure in the spatial pattern generation of a CLC-laser.

2. Samples and experimental set-up

As the matrix of a steroidal cholesteric, we used a three-component mixture containing 40% cholesteryl oleate, 35% cholesteryl pelargonate, and 25% cholesteryl chloride with the temperature alteration of a helical pitch ≈ 3 nm/degr. The matrix cholesterol was doped with the phenolenone dye F490 with weight concentration 0.3%. The structural formulas of cholesterol esters and dye are presented in Fig. 1. This mixture has a left twisting spiral and the maximum selective reflection (SR) band in 600 nm. The thickness of the dye- doped cholesteric in the oriented planar texture was 45 μm.

A peculiarity of the mixture of cholesterol esters is its high viscosity. Therefore, the relative shift of substrates in the direction of rubbing was used for the orientation in addition to the standard method of rubbing the orienting layer on the substrate [16].

The planar texture was created by means of the standard technique. The technology includes the rubbing of the glass or quartz substrates coated with ITO layer and polyimide lacquer [16] and their further mutual shift in the rubbing direction at the phase transition temperature after the filling of the sample. In the manufacture of textures with orthogonal director orientations on substrates, instead of their shift after the sample cooling, we used a small reversal of one of the substrates at an angle of 20° and back.

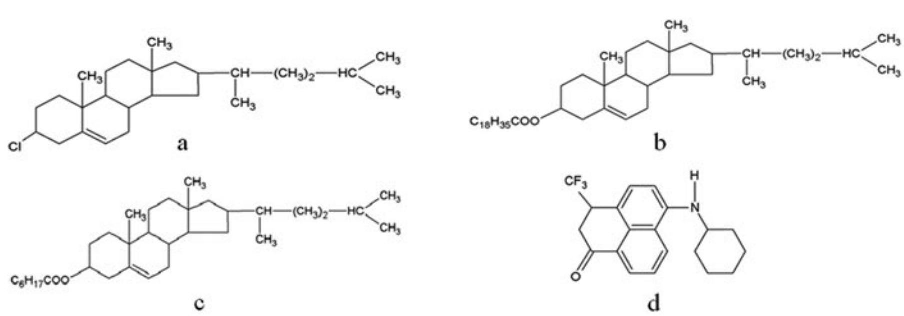

Figure 1. Chemical structure of used materials: a - cholesteryl chloride; b - cholesteryl pelargonate: c-cholesteryl oleate: d - phenalenone dye F490.

The lasing characteristics of dye-doped cholesterics were studied on the typical experimental setup. The optical pumping of the dye-doped CLC was carried out by the second harmonic ($\lambda = 530$ nm) of a Q-switched laser on neodymium glass operating in a slow pulse repetition rate mode with the pulse duration $\approx$20 ns. The second harmonic radiation was focused by a lens with a focal distance of 21 cm on the sample of the dye-doped CLC on a spot 0.5 mm in diameter.

A maximal power density of the second harmonic radiation was $\approx$ 27 MW/cm^2 and was attenuated by neutral filters. The lasing spectra of the dye-doped CLC-laser corresponding to each pumping pulse were optically imaged in a focal plane of a spectrograph with an inverse dispersion of 0.6 nm/mm and then displayed by the video camera on a PC monitor.

3. Results and discussion

The study of transmission spectra of steroid cholesteric formed by cholesterol esters with low birefringence shows a clear dependence of the selective reflection bandwidth on the quality of a planar texture. We note that a better orientation is observed on substrates coated with an additional layer of transparent ITO [16]. The quality of planar texture affects the transmission spectra, where significant differences are observed, as is shown in Fig. 2. Here, the transmission spectrum of the oriented samples with a planar texture of the cholesteric based on a three-component mixture of cholesterol esters with the above-mentioned percent composition at the use of orienting substrates with ITO is presented. For planar textures created with the use of glass and quartz substrates coated with the ITO layer and the layer of polyimide lacquer, the half-width of the SR-band $\approx$ 22 nm (Fig. 2). If the substrates with ITO forming a planar texture are absent, its quality becomes worse markedly, which is manifested in the broadening of the SR-band by more than 10% and a decrease in the magnitude of diffraction reflection (Fig. 3).

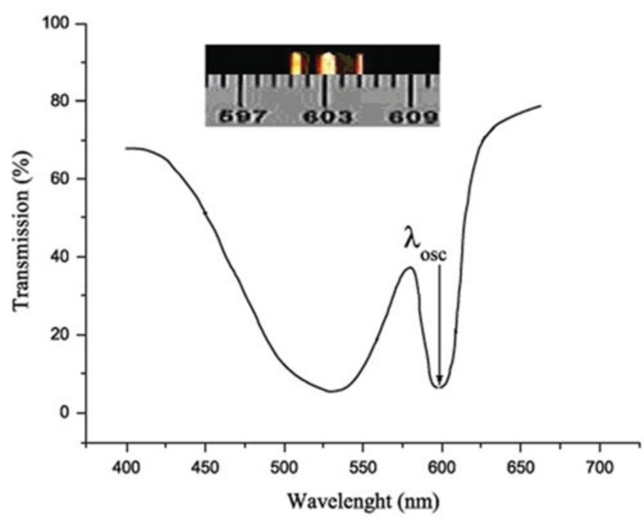

Figure 2. The transmission spectrum of a qualitative planar texture of the dye-doped CLC formed by substrates with ITO. The arrow shows the lasing spectrum location. The insert represents the lasing spectrum in such structure. The peak near 600 nm is the maximum of the CLC selective reflection band.

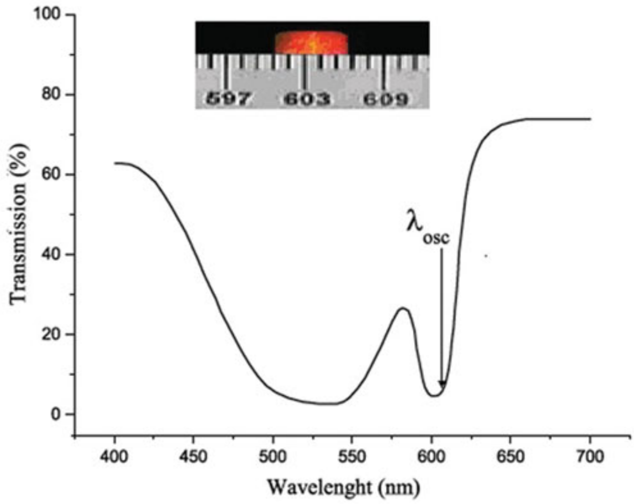

Figure 3. The transmission spectrum of a planar texture of the dye-doped CLC formed by substrates without ITO. The arrow shows the lasing spectrum location. The insert represents the lasing spectrum in such structure.

The study of lasing spectra in steroid cholesteric shows a clear correlation with the quality of a texture. As shown in Fig. 2 (spectrum above), the lasing spectrum discovers three longitudinal modes for a high-quality texture with a narrow SR-band for parallel mutual orientation of directors on substrates. In such a structure, the lasing spectrum is located at the center of the SR-band, which corresponds to the coupled wave model of the lasing in a periodic structure [10]. An increase of the pumping intensity is not followed by an increase of the number of longitudinal modes until the intensity level, at which the destruction of a sample occurs.

It should be noted that the presence of the ITO layer affects only the ordering of a planar texture and is manifested in the SR-band width and the Bragg diffraction efficiency in it. The lasing outside the SR-band under the conditions of a nonselective resonator created by transparent electrodes with coefficients of reflection mirrors $R = 8 \div 10\%$ did not arise in the whole range of excitation intensities up to the destruction of the sample.

The presence of only three longitudinal modes in the lasing spectrum in the entire range of excitation indicates a high selectivity of the helical periodic structure, which is manifested in the suppression of longitudinal modes with higher indices. This is consistent with the model of coupled waves [10] and the theoretical model of distributed feedback helical laser, which was developed in [17].

Figure 3 (inset top) shows the lasing spectra with the same active medium components as in Fig. 2, but the planar texture is formed on the substrates without ITO. As can be seen, a slight 3-nm shift of the lasing spectrum from the center to the long wavelength edge of the SR-band appears, and the structure with discrete modes is changed by a diffuse broad band. This is followed by the about one order increase of the lasing threshold intensity.

A conventional planar oriented dye-doped cholesteric laser emits a beam with high angular divergence, which is caused, in the first turn, by a small thickness of the active medium. As shown in Fig. 4b, the spatial emission pattern contains, along with the intensive central zone, the ring structure. The number of rings depends on the excitation intensity. The same figure (Fig. 4a) presents the dependence of the angular divergence on the pumping intensity for the layer thickness of 45 μm. As can be seen, the angular divergence increases sharply within 80 pumping thresholds up to 17°.

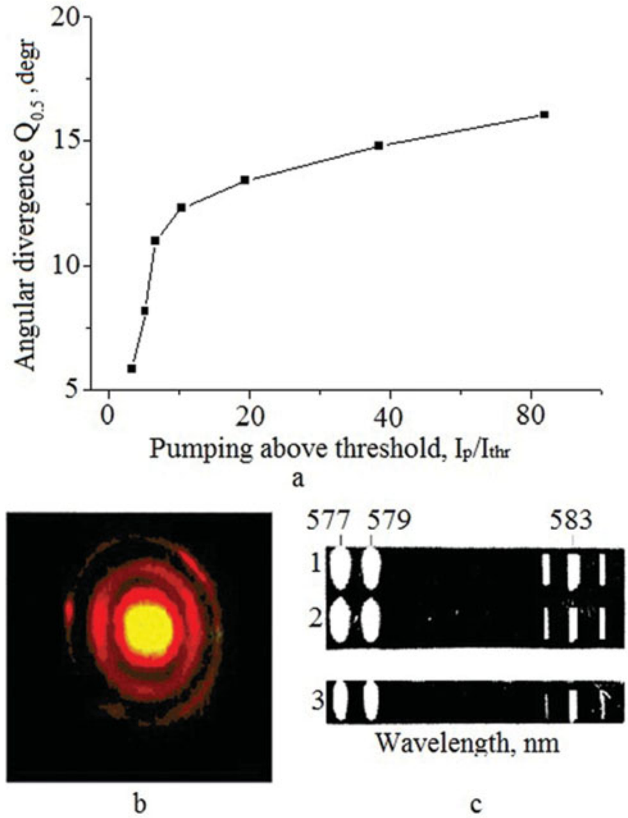

Figure 4. The angular divergence and lasing spectra of a CLC-laser at the parallel orientation of directors on substrates; a – dependence of the angular divergence on the level of excitation; b - ring structure of the laser beam; c - lasing spectra in the central core (1), first (2) and second (3) rings at Ip / I thr $\approx$ 12; 577 and 579 nm - reference lines of the mercury doublet.

We note that two nonlinear effects would be the possible mechanisms of the appearance of a ring structure in the cross-section of the beam of a CLC-laser, which is generated at a pulse nanosecond pumping. They are a rotation of the director of a liquid crystal along the vector of polarization of a light wave and the formation of a thermal lens [18]. However, the studies of the lasing spectra with a resolving power of $\sim$0.5 nm in the central core and two first rings showed their practical invariability (Fig. 4c), which exclude the influence of nonlinear effects.

As distinct from work [19], where the ring structure of the beam of a CLC-laser was explained by the diffraction due to a small excitation zone, we explain the ring structure by the lasing of modes propagating at discrete angles to the resonator axis. An analogous effect is observed also in solid-state lasers [20]. Such explanation was confirmed in work [21], where the lasing wavelengths for rings were studied with high resolving power, and the shift of at least 1 nm was registered for high-order rings as compared with the central zone. A change in the lasing wavelength along the cross-section of the beam does not correspond to the diffraction model of rings.

The typical cholesterics have a high viscosity and are oriented mainly by a shift. Earlier, we established [22] that, at the application of textures with orthogonal orientation of directors on substrates, it is possible to get the one-mode lasing for a CLC-laser, which is not realized under conditions of the ordinary planar texture (with parallel directors on substrates) due to

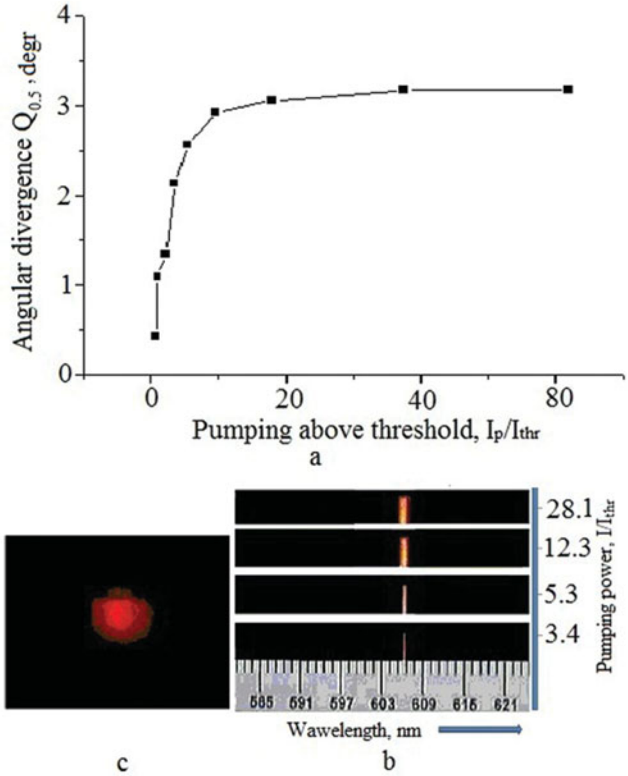

Figure 5. The angular divergence and lasing of a CLC-laser at the orthogonal orientation of directors on substrates; a – dependence of the angular divergence on the level of excitation; b - lasing spectra at various excitation intensities; c - section of the laser beam.

the closeness of the lasing thresholds of longitudinal modes. In order to improve the characteristics of a CLC-laser, we studied the spectral and spatial characteristics of such texture.

We used the quartz substrates with a layer of polyimide lacquer, whose thickness was$\sim$1 μm. The technology included the watering of substrates by a 10% solution of polyimide, standard procedure of rubbing, orthogonal arrangement of the directions of rubbing on substrates, and turn of the upper substrate relative to the lower one by an angle of $\sim$ 20° with the return to the initial position before the pressurization of the layer.

For such CLC texture, we observe certain specific features in the lasing characteristics.

As shown on Fig. 5b, the lasing spectrum of such structures contains only the lowest longitudinal mode. The lasing spectrum width decreases by more than one order, reaching the tenth fractions of a nanometer. The lasing threshold in such structure is higher by more than order, and the active area (anomalous zone) is spatially localized in a domain up to 2 cm in width, which bounded by the disclination lines over the whole width of the specimen (3 cm). As can be seen from Fig. 5b, the single-mode lasing is realized in this area in the range of excitation intensities up to the fracture of a specimen. One more peculiarity of the lasing spectrum in the anomalous zone is a shift of the lasing spectrum to the long-wave region by more than 3 nm, as compared with the lasing spectrum outside the anomalous zone.

The spatial pattern of a lasing for the mutually crossed orientations of the surface layers of molecules on substrates does not manifest any ring structure. There is only the central core, and its angular divergence is not higher than about 3 degrees. In Fig. 5a, we show the dependence of the angular divergence on the pumping power as well. Outside of the anomalous zone on the same specimen, we observe the typical ring structure of the laser beam.

Similar features in the lasing characteristics were already observed for viscous mixtures of cholesterol esters. But, for nonviscous induced cholesterics, they were not observed. The study of the influence of various substrates showed that a strong selection of both longitudinal and circular modes is realized only with quartz substrates.

A significant increase of the lasing threshold for the texture of steroidal CLCs with orthogonal orientation of the surface layers of molecules can indicate that the lasing involves only a certain limited thickness of the active layer region, which can testify to the presence of a defect in such structure.

The theory [23] forecasts the possibility of a one-mode lasing under conditions of a defect in the spiral structure of a CLC, including the jump of a phase, which can be formed at the turn of orienting substrates. However, we have not registered any dips in the selective reflection band, which is forecasted by this theory.

4. Conclusions

1. It is demonstrated experimentally that, at the perpendicular orientation of directors on CLC orienting substrates, the single-mode lasing under a multiple excess of the threshold can be achieved. This method improves the spectral purity and the angular radiation divergence of a CLC-laser considerably.
2. The interrelation between the spectral and spatial characteristics of the lasing in steroidal CLCs can be formulated as follows: the ring structure of the laser emission is related to the lasing of many discrete modes and disappears under the single-mode lasing, which disagrees with the diffraction model of formation of rings.

References

[1] Ilchishin, I. P., Tikhonov, E. A., Tishchenko, V. G., & Shpak, M. T. (1980). *JETP Lett.*, *32*, 24.
[2] Chanishvili, A., Chilaya, G., Petriashvili, G., Barberi, R., Bartolino, R., Cipparrone, G., Mazzulla, A., Gimenez, R., Oriol, L., & Pinol, M. (2005). *Appl. Phys. Lett.*, *86*, 051107.
[3] Taheri, B., Munoz, A. F., Palffy-Muhoray, P., & Twieg, R. (2001). *Mol. Cryst. Liq. Cryst. 358*, 73.
[4] Muñoz, A., McConney, M. E., Kosa, T., Luchette, P., Sukhomlinova, L., White, T. J., Bunning, T. J., & Taheri, B. (2012). *Opt. Lett. 37*, 2904.
[5] Mykytiuk, T. V., Ilchishin, I. P., Yaroshchuk, O. V., Kravchuk, R. M., Li, Y., & Li, Q. (2014). *Opt. Lett. 39*, 6490.
[6] Lin-Jer, Chen, Jia-De, Lina, & Chia-Rong, Lee (2014). *J. Mater. Chem. C*, *2*, 4388.
[7] Dolgaleva, K., Wei, S.K.H., Lukishova, S. G., Chen Sh.H., Schwertz, K., & Boyd, R. W. (2008). *J.Opt. Soc. Am. 25*, 1496.
[8] Ilchishin, I. P. (1996). *Bulletin Russian Academy of Sci. Phys. 60*, 494.
[9] Ilchishin, I P., & Tikhonov, Eu. A. (2015). *Progress in Quant. Electronics. 41*, 1.
[10] Kogelnik, H., & Shank, S. V. (1972). *J. Appl. Phys. 43*, 2327.
[11] Kopp, V. I., Fan, B., Vithana, H. K. M., & Genak, A. Z. (1998). *Opt. Lett. 23*, 1707.
[12] Schmidtke, J., Stille, W., Finkelmann, H., & Kim, S. T. (2002). *Adv. Mater. 14*, 746.
[13] Wei, S.K.H., Chen Shaw, H., Dolgaleva, K., Lukishova, S. G., & Boyd, R. W. (2009). *Appl. Phys. Lett. 94*, 041111.
[14] Haase, W., Podgornov, F., Matsuhisa, Y., & Ozaki, M. (2007). *Phys. stat. sol. (a)*, *204*, 3768.
[15] Takanishi, Y., Tomoe, N., Ha, N. Y., Toyooka, T., Nishimura, S., Ishikawa, K., & Takezoe, H. (2007). *Jap. J. Appl. Phys. 46*, 3510.
[16] Denisov, Yu. V., Kizel, V. A., & Sukhenko, E. P. (1976). *JETP*, *71*, 679.

[17] Preiswerk, H. P., Lubanski, M., Gnepf, S., & Kneubuhl, F. K. (1983). *IEEE J. Quant. Electronics*, *QE-19*, 1452.
[18] Ilchishin, I. P., Tikhonov, E. A., & Shpak, M. T. (1988). *Ukr. Phys. J. 33*, 10.
[19] Kopp, V. I., Zhang, Z.-Q., & Genack, A. Z. (2003). *Prog. Quant. Electron.*, *27*, 369.
[20] Stoicheff, B. P., Szabo, A. (1963). *Appl. Opt.*, *2*, 811.
[21] Palto, S. P., Shtykov, N. M., Umansky, B. A., Barnik, M. I., & Blinov, L. M. (2006). *Opto-Electronics Review*, *14*, 323.
[22] Gryshchenko, S. V., & Ilchishin, I. P. Single-mode oscillations in DFB-laser based on cholesteric liquid crystal. *Presented at the X International Conference on Laser Optics*, St.-Petersburg, Russia, June 26–30, 2000.
[23] Belyakov, V. A., & Semenov, S. V. (2011). *JETP*, *112*, 694.