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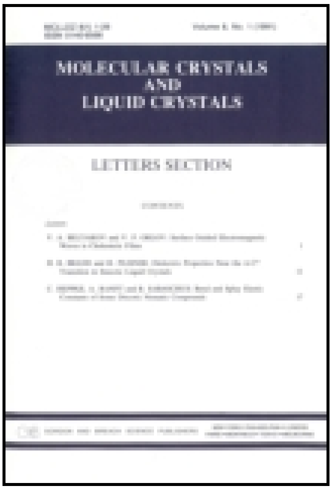

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I. P. Ilchishin [a] & T. V. Mykytiuk [a]
[a] Institute of Physics of the NAS of Ukraine , 46, Nauky Prosp, Kyiv , 03680 , Ukraine

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Lasing Spectra of Dye Doped Cholesteric Liquid Crystals Under Reversible Phototuning of the Frequency

I. P. ILCHISHIN AND T. V. MYKYTIUK*

Institute of Physics of the NAS of Ukraine, 46, Nauky Prosp., Kyiv 03680, Ukraine

We investigate the lasing frequency phototuning in a distributed feedback (DFB) laser based on cholesteric liquid crystals (CLC). To improve the CLC planar texture quality, we use new materials such as nematic ZhK-654 and an analog of cholesteryl oleate as a twisting additive. The selective excitation of the cis*-form molecules of the azoxy-nematic is carried out with the use of a combination of an interference filter with maximum transmission at 436 nm and a cut-off filter. Using the orienting substrates with SnO_2 allows us to reduce the threshold pump power by more than an order of magnitude. The proposed combination of filters for the selective excitation enables us to align the ranges of lasing wavelengths in both directions.*

Keywords Cholesteric liquid crystal; distributed feedback laser; lasing spectra; phototuning

1. Introduction

Dye-doped CLCs with natural periodic structure are suitable structures for lasing. On the basis of such materials, mirrorless microlasers have been created [1]. The absence of an external cavity, arbitrariness of the active medium cross-section, and low lasing thresholds indicate their possible use in the information displays with high brightness. The main method used for the lasing frequency tuning in CLC-based DFB lasers is based on a helical pitch variation with a change of the temperature [1]. The low tuning speed and the need of a CLC thermostabilization hinders a broad application of such lasers. Therefore, the search for new and more efficient methods of lasing frequency tuning in CLC is an important problem.

It is known that, in some types of induced CLC based on nematic liquid crystals (NLC) with chiral dopants (CDs), one can change the molecular conformations of these dopants by UV irradiation with the corresponding changes in their helical twisting power. This type of CLC, including those with non-mesogenic CDS, because of the presence of different photoisomer forms of molecules and the easiness of their phototransformation [2], can ensure a variation of their helical pitch under the excitation of a certain molecular stereoisomeric form.

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*Corresponding author, T. V. Mykytiuk. Phone: +38-044 5250929, Fax: +38-044 5251589. E-mail: taras.mykytiuk@gmail.com

Earlier, the effect of changes in the dopant helical twisting power due to the transition from the *trans*- to *cis*-conformation, which is accompanied by a variation of the helical pitch, was used for the holographic recording [2], as well as for the frequency lasing control of CLC-based DFB-lasers [3–5]. Since the CDS were used with the very close absorption bands of *trans*- and *cis*-conformations [5], this method of phototuning did not allow one to selectively irradiate the mentioned molecular conformations and to control their composition in the irradiation zone. Thus, the frequency tuning can be only unidirectional with increasing the *cis*-conformation concentration under irradiation and decreasing the helical twisting power of a dopant. In this case, the lasing line continuously shifts toward longer wavelengths. The reverse process, i.e., the restoration of the initial stereoisomer composition, occurs spontaneously during 17–20 h and is practically independent of the external factors (temperature, electric field, etc.) [5].

Recently, new ways have been proposed for the realization of a controlled reversible phototuning of the helical pitch in CLC. To achieve this, it was proposed [6–7] to use nematics on the basis of azoxy compounds (azoxy nematics) with the absorption bands of stereoisomers located in such a manner that the irradiation with light wavelengths above ∼410 nm results in a selective irradiation of only a single *cis*-conformation. Such location of the absorption bands allows their selective excitation and, by that, the reversal direction of lasing frequency tuning. Recently, a possibility of lasing in such materials was reported in [8]. In [6–7], it was proposed to use a mixture of azoxy-nematics ZhK-440 for the reversible frequency phototuning in CLCs. It was shown in [9] that the poor quality of the planar texture of CLCs leads to a large increase in the lasing threshold and a disparity in the frequency tuning ranges in both directions.

Therefore, our work was aimed at the study of new materials that would ensure a better quality of the planar texture of CLCs and at the search of ways to reduce the lasing thresholds and to align and to extend the frequency phototuning ranges in both directions.

2. Samples and Experimental Set-Up

As a matrix, for the realization of a reversible phototuning, we used CLC composed of 36% Tekon-20 (analogue of cholesteryl oleate, Institute of Single Crystals of the NAS of Ukraine) + 64% nematic ZhK-654, which incorporates 66% azoxy-nematic ZhK-440 (2:1 mixture of 4-*n*-butyl-40-methoxyazoxybenzene and 4-*n*-butyl-40-heptanoylazoxybenzene, NIOPIK, Russia). CLC was doped with phenolenone dye F490 (NIOPIK, Moscow, Russia) with a weight concentration of 0.3-0.4%. Such compositions have been used previously for studing the induced CLC lasing and has showed a relatively low threshold pumping intensity [10]. The planar texture was produced by the known method of liquid crystal (LC) orientation with the surface rubbing of quartz substrates coated with a layer of SnO_2 and a polyimide lacquer. The UV-vis absorption spectra of the CLC were measured at room temperature by a spectrophotometer SF-20 (LOMO, St.-Petersburg). The fluorescence spectrum of a dye in CLC is measured by a spectrometer MPF-4 “Hitachi”. The maximum of the selective reflection (SR) band was chosen to be superimposed with a fluorescence maximum in the spectral region of about 600 nm. For the UV irradiation, we used a low-pressure Hg lamp of the type DRK-120 with air cooling (a radiation power of 120 W, stabilized discharge current). To reverse the light irradiation, we used the same lamp with filter ZhS-11 and an interference filter with the maximum transmittance at 436 nm and a bandwidth of 20 nm.

The optical pumping of the doped CLC was carried out by the second harmonic ($\lambda = 530$ nm) from a Q-switched Nd^{3+} laser operating in a slow pulse repetition rate mode

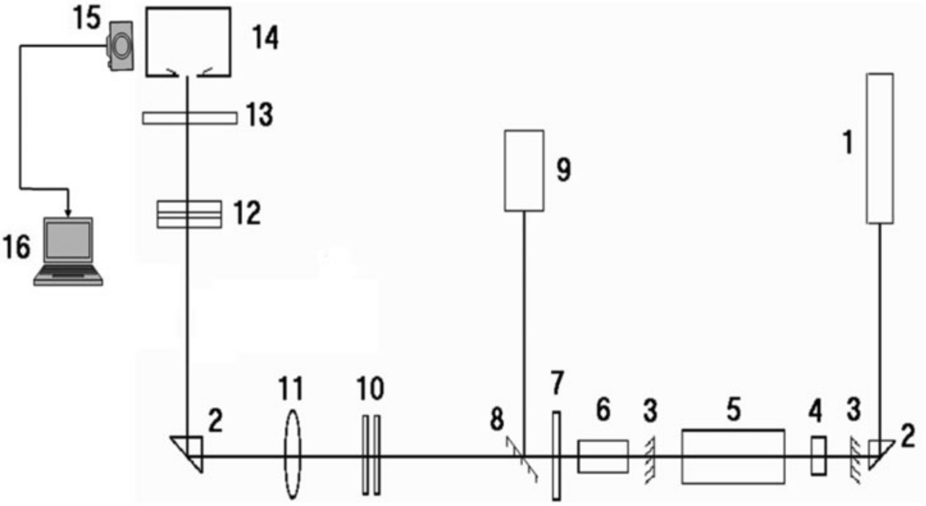

Figure 1. Schematic of a laser setup: 1 – He-Ne laser, 2 – rotating prism, 3 –resonator mirrors, 4 – Q-switched modulator, 5 – active element of a solid-state laser, 6 – frequency doublers, 7 – filter, 8 –half mirror, 9 – calorimeter, 10 –neutral filters, 11 – lens, 12 – dye-doped CLC, 13- filter, 14 – spectrograph, 15 – video camera, 16 –computer.

with the pulse duration $\approx$ 20ns (Fig. 1). The second harmonic radiation was focused with a lens of a focal distance of 21 cm on the doped CLC sample in a spot with a diameter $\approx$1.0 mm. The maximal second harmonic power density was $\approx$ 27 MW/cm^2 and attenuated by neutral filters. The lasing spectra of the dyed CLC corresponding to each pumping pulse have been optically imaged in a focal plane of a spectrograph with an inverse dispersion of 0.6 nm/mm and then displayed in a video camera on a PC monitor.

3. Results and Discussion

As is shown in Fig. 2, azoxy-nematic ZhK-654 in the induced CLC, which is used instead of ZhK-440, improves the optical properties of planar textures that can be seen from the intensity spectrum of SR in the reverse regime (Fig. 2, curve 3). The application of the proposed combination of filters to the selective excitation of *cis*-form azoxy-nematic molecules allows us to eliminate the difference between the phototuning bands in the short- and long-wave directions. After 20 min of UV irradiation of the induced CLC-based azoxy-nematic ZhK-654, we got the tuning in the forward direction by 34 nm (Fig. 2, curve 2). This is lower than that in the same experiment with the CLC based on pure ZhK-440 [10]. The growth of the reversal time to 60 min (Fig. 2, curve 3) can be explained by the low-intensity selective excitation by using the filters mentioned above and a low level of absorption of the *cis*-conformation of an azoxy-nematic molecule in the area of their self-absorption [10].

An extraordinary effect occurs in the planar texture after the irradiation through a combination of filters. As is shown in Fig. 2, curve 3, after the reverse shift of the SR band, its intensity increases by more than 20%. The band intensity increase can be attributed to a better ordering of azoxy-nematic molecules in the planar texture of CLC in the transition from the *cis*- to *trans*-form. The measurements of the transmittance spectra of CLC in a long time have shown that this effect is unstable. In 24 h, the transmittance level relaxes to that shown in Fig. 2, curve 1. Thus, the combined effect of surface adhesion forces

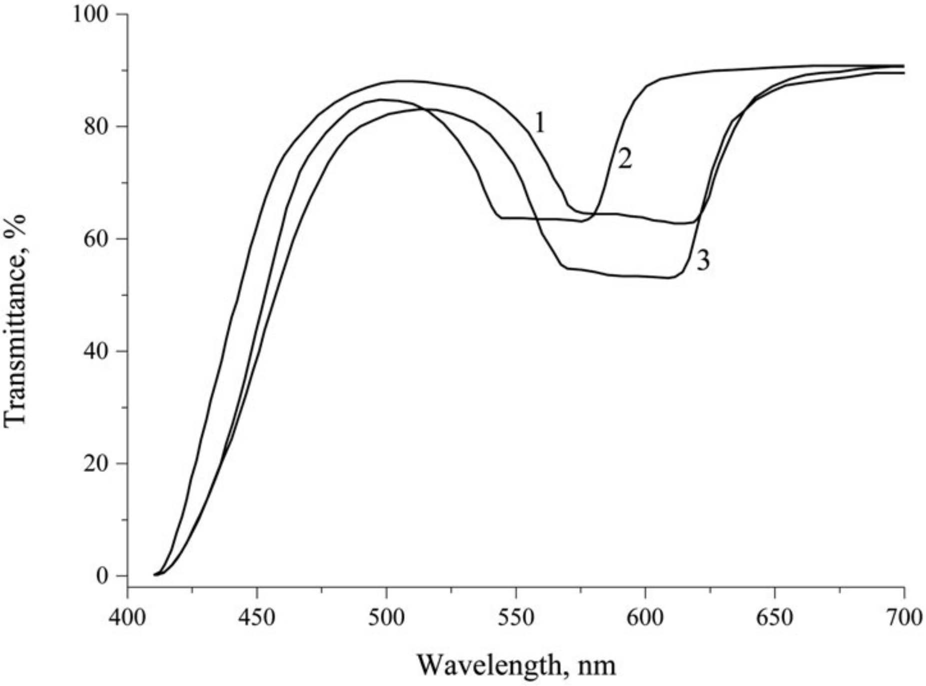

Figure 2. Transmittance spectra of the liquid crystal mixture 36% Tekon-20 + 64% ZhK-654; 1- under UV irradiation; 2 – 20-min UV irradiation; 3 – 60-min irradiation with filter ZhS-11 + interference filter. The layer thickness is 40 μm.

and elasticity forces in the liquid crystal leads to the spontaneous establishment of the equilibrium position of the *trans*-forms of azoxy-nematic molecules and the corresponding change in optical characteristics of the CLC planar texture. The clarification of the nature of this effect requires more detailed studies.

It is known [9] that the induced CLC on the basis of a mixture of nematic ZhK-654 and cholesterol ester as a twisting impurity in creating a DFB laser can be activated by phenolenone dye F 490 with a large Stokes shift (60 nm) between the maxima of the absorption and fluorescence spectra. This activator is more promising for phototuning the laser frequency than pyrromethene dye PM597 with a Stokes shift of 40 nm previously studied in [10]. At the activation of CLC induced by such dye and the phototuning due to the *trans-cis* photoisomerization, the tuning time was the same as that for pure CLC. At the reversing, i.e., at the *trans-cis* photoisomerization, the time of the selective exposure to attain the full reverse of a helix pitch increases, as compared with that of pure CLC, to 100 min. This is due to the weakening of the selective irradiation by a combination of filters due to the dye absorption in this spectral range.

At the pulsed excitation of a sample of the induced CLC planar texture created on the basis of the above-described mixture of the azoxy-nematic and a twisting dopant Tekon 20 and a thickness of the active layer of 40 micrometers, the threshold pump intensity is almost by an order of magnitude lower than that with the use of nematic ZhK-440 [9]. A DFB laser based on this mixture generate at a wavelength of 619 nm, which corresponds to the long-wavelength edge of the SR band. When the sample is irradiated with UV irradiation from a mercury lamp for 20 min at a distance of 15 cm from of the CLC sample, then the

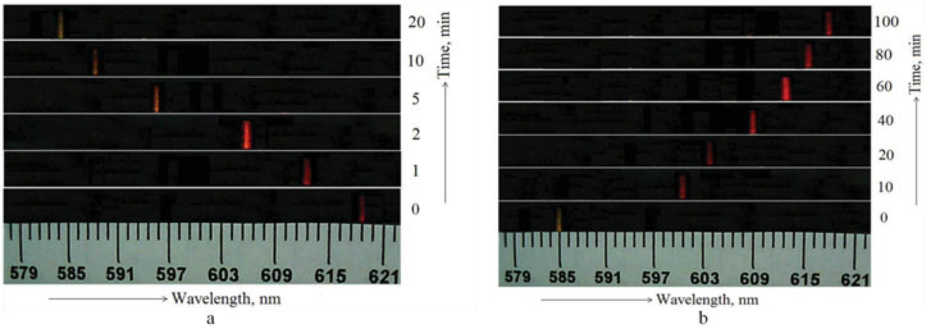

Figure 3. Phototuning in the forward (a) and reverse (b) directions of lasing spectra of induced CLC based on the mixture 36% Tekon-20 + 64% ZhK-654. The CLC layer thickness is 40 μm. The wavelength scale interval is 1.2 nm.

lasing spectra of a DFB- laser is shifted into the short-wave region to 585 nm (Figs. 3 and 4, curve 1). The phototuning range of a lasing line in the short-wave direction is 34 nm, which corresponds to the CLC helix pitch.

Figure 3 represents the tuning lasing spectra under the UV-irradiation (a) and the irradiation with a mercury lamp through a selected combination of filters (b), which cuts

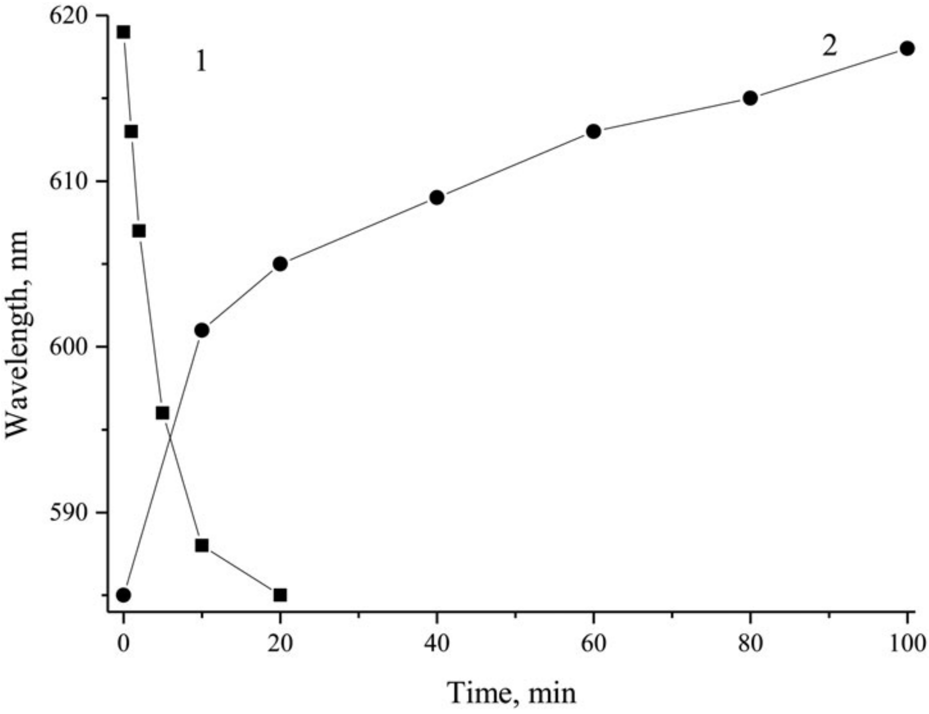

Figure 4. Lasing wavelength tuning of dye F 490 in the induced CLC on the basis of nematic ZhK-654 as a function of the UV exposure time (1) and reverse phototuning (2) after irradiation with filter ZhS-11 + interference filter under the same conditions. The layer thickness is 40 μm.

the spectrum of this lamp to a narrow lane. It corresponds to the absorption of only the *cis*-form of a azoxy-nematic molecule. The excitation intensity was chosen so that, under the tuning in the maximum fluorescence dye area at 590 nm, at most one of the lowest longitudinal modes appears in the laser spectrum. As is seen from Fig. 3, the phototuning ranges in both directions are the same at a width of the spectrum of single-mode lasing of 0.3-0.5 nm and equal to 34 nm.

Figure 4 (curve 2) shows an increase in the time tuning of a lasing frequency of the induced CLC in the reverse direction. The nature of the curve shows that the process of *cis-trans* isomerization of molecules LCD 654 is nonlinear relative to the illumination intensity. Moreover, the saturation of this transition does not occur within 100 min. This results in the possibility to expand the range of phototuning of the lasing frequency. The only option of reducing the reverse process duration is the irradiation of azoxy-nematic molecules with spectral intensity by orders of magnitude greater than a mercury lamp and with a narrow band at 412–415 nm.

4. Conclusions

The use of azoxy-nematic ZhK-654 instead of ZhK-440 can significantly reduce the threshold excitation intensity and allow one to obtain the same frequency ranges of tuning the lasing frequency in CLC in both directions.

The increase in the selective reflection intensity at reversing the helix pitch of CLC is caused by a better ordering of its molecules at the transition from *cis*- to *trans*-conformation. However, the instability of this process and the return of a SR intensity to the initial level during a day require more detailed studies.

Reducing the lasing frequency phototuning time in such material in the reverse direction can be achieved by a selective irradiation with sources with high spectral intensity in the interval 412–415 nm, for example, with the laser diodes.

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