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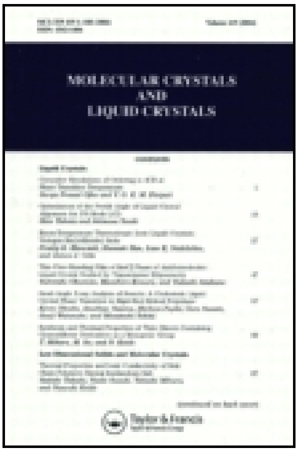

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Detecting of the Structure Distortion of Cholesteric Liquid Crystal Using the Generation Characteristics of the Distributed Feedback Laser Based on It

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DETECTING OF THE STRUCTURE DISTORTION OF CHOLESTERIC LIQUID CRYSTAL USING THE GENERATION CHARACTERISTICS OF THE DISTRIBUTED FEEDBACK LASER BASED ON IT

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Abstract Threshold and polarization characteristics as well as temperature dependence of a two-band generation of laser with distributed feedback (DF) based on induced cholesteric liquid crystals (CLC) were studied. Difference between threshold excitation intensities of these bands of generation points at the existence of two phase gratings in the helical structure of CLC. It is verified by the difference in the temperature behaviour of the wavelengths of the central lines of the bands.

INTRODUCTION

Natural periodic structure of CLC doped with lasers dye permits the design of compact lasers with CLC-based DF.[1] Temperature control of the CLC pitch is a simple and reliable way to tune the generation frequency of such a laser.[1] As another laser coherent radiation is used as an excitation source CLC-based one could be spoken about as an adapter for laser radiation transformation. Nevertheless, threshold values of generation in CLC helical structure allows the use of incoherent pulse ($\tau_p \leq 100$ ns) radiation sources as well.

Along with a practical use of dye-doped CLC as a source of a tunable laser radiation there is an obvious possibility to study a supra-molecular CLC-structure with the help of these lasers. This might yield a useful knowledge for their various applications, for instance, for information displays, etc.

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Even in early experiments dealing with laser generation in steroid CLC[1] such a laser was established to possess a higher selectivity then DF-laser (DFL) based on an ordinary linear periodic structure.[2] Lately this conclusion was also made in a theory.[3] High spectral selectivity of a CLC-based DFL consists in the fact that only three generation lines, central (Bragg) one and two side-ward longitudinal modes, are observed through the overall excitation intensity range up to the heat destruction of the helix.[4] According to the physical concept of DFL[2] existence of any additional lines or bands in DFL generation spectrum, besides the three mentioned above, obviously points to either the cholesteric pitch irregularity or a periodic helical distortion. Such structure deformity appeared to be actually invisible in the selective reflection (SR) spectrum because of a strong diffractive light scattering.

As was established earlier[5] on the base of laser generation spectra there is a harmonic distortion of the helical structure induced by twisting dopants in a nematic liquid crystal (NLC). The recent model of an induced CLC[6] predicts the existence of two incommensurate periods in helical structure provided by the definite ratio of K_{11} and K_{22} elastic constants. These periods are originated by static solitons in the CLC-director orientation. Two-band generation in induced CLC[7] seems to be a qualitative confirmation for the model.[6] The comparison of the theory and experimental data received on the induced CLC,[7] mixture 4-methoxybenzylidene-4'-butylaniline (MBBA) and a non-mesogenic twisting dopant, could be considered as the first quantitative treatment. More concise examination of the model[6,8] is difficult because of the inaccuracy of elastic constants determination (especially for mixtures of NLC with CLC, that are very viscous) and its phenomenological character.

In the present paper we consider the procedure for identification of harmonic distortions of the helical structure in mixture of nematic liquid crystal with CLC via generation spectra of laser based on it. We display our results on generation threshold for the two bands, their polarization, temperature dependence of generation spectra and their relation to the analogous dependence of SR band. They are also discussed from the point of view of accordance of features of each DF-laser generation band to a certain period of the helical structure of induced CLC.

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EXPERIMENT, RESULTS AND DISCUSSION

Multicomponent mixture ZhK-654 was used as a nematic matrix with adding of 36-37 weight percentage (wt.%) of cholesteryloleate (CO). This compound was activated by a generating dye of phenalenone type (0.3 wt.%). Planar CLC structure was placed between glass plates covered with SnO_2 transparent electrodes and a rubbed layer of polyvinyl alcohol. Film thickness of 30 μm was given by calibrated spacers. Such film thickness provided good planar texture and threshold excitation intensity of DF-laser was not greater then 1 MW/cm^2. Selective reflection spectra of pure and dye-doped CLC were obtained in unpolarized light with the help of spectrophotometer SF-20. Fluorescence spectra of the dye were obtained on the MDF-4 Hitachi spectrometer. A second harmonic of the radiation of Q-switched Nd^{3+}-glass laser, working in a single pulse regime (λ=530 nm), focused to a 0.3 mm diameter spot was used for excitation. Exciting beam passed along the normal to the cell, that was placed in front of the spectrograph forward slit, and was cut by a colour filter. Generation spectra were fixed on a photo film in the spectrograph with 6 Å/mm dispersion. Excitation intensity was controlled by neutral filters and mounted up to 40 MW/cm^2. Heating of the cell was provided by the electric current passed through low-ohm transparent electrodes on the outer sides of glass substrates. Temperature control of ±0.5 °C accuracy was measured by a thermocouple.

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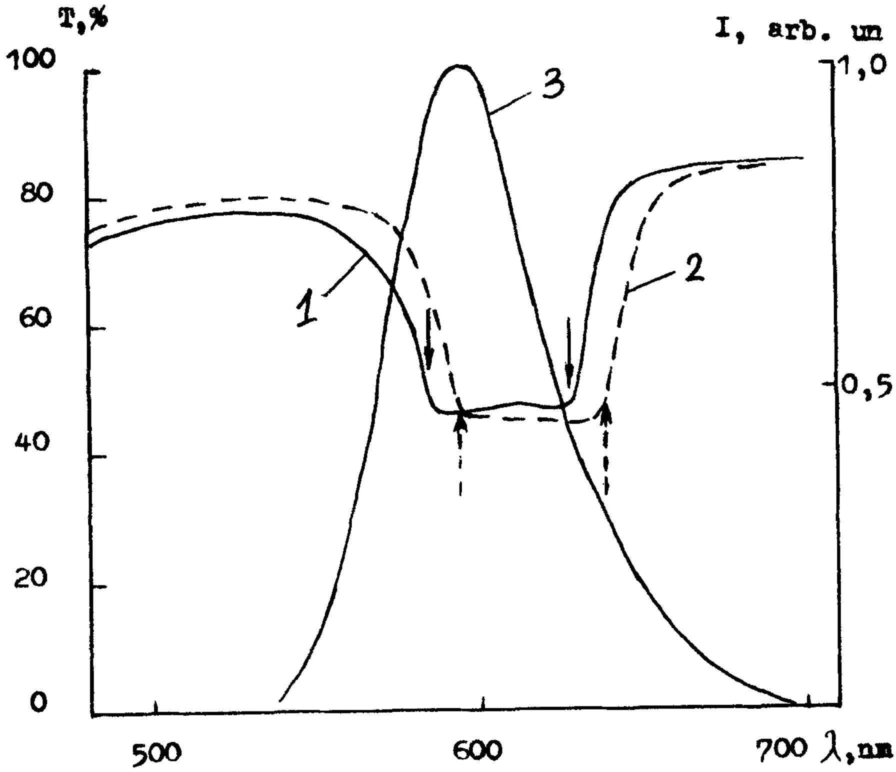

FIGURE 1 1, 2 - light transmission spectra of the dye-undoped sample I and II respectively in unpolarized light, 3 - fluorescence spectrum of the doping dye in CLC.

Transmission spectra of two cells filled by the mixture ZhK-654 + CO with CO percentage of 37 wt.% for sample I (curve 1) and 36 wt.% for sample II (curve 2) are shown on Fig.1. After dye-doping of these compounds two-band DF-laser generation (each band was a triplet) was registered. Bragg (central) wavelengths (λ_B) of the

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triplets are indicated in Fig.1 with arrows. At the temperature of 18 $^{\circ}C$ values of λ_B for sample I were 582 and 626 nm, for sample II they were 591 and 637 nm. As could be seen from the spectrum of the dye fluorescence in CLC (Fig.1, curve 3), which agrees with its gain spectrum, conditions of generation in these cells are strongly different for both bands. Threshold excitation intensities for short-wavelength band appeared to be 4.5 times greater then that in the band with λ_B=626 nm. Taking into account a small Stocks shift (60 nm) of dye absorption and fluorescence bands, an increase of the generation threshold in the band with λ_B=582 nm could be explained by a high loss level caused by absorption. However, for the sample II generation band with λ_B=591 nm coincides virtually with the fluorescence band maximum and re-absorption losses are minimal here. Nevertheless, its generation threshold was registered to be the same as for the band with λ_B=637 nm, where fluorescence intensity is 2.8 times less.

According to the principle of DF-laser operation[2] the existence of two bands of generation testifies undoubtedly about two periods in a periodic structure. Data on the investigation of steroid CLC-based DF-laser[1,4] are in agreement with the general theoretical conclusions,[2] except the higher spectral selectivity of generation. On good planar CLC textures, without a helical period gradient as well as areas differing in CLC pitch, the wavelength of the central line of generation λ actually coincides with the selective reflection maximum λ_B. The planar texture of induced CLC is of sufficient quality too: the local colour irregularities are not revealed either by transmission measurements or by optical microscope. Just using the laser generation spectra of doped CLC it is possible to detect these two near-by periods.

An equality of the threshold excitation intensities of both DF-laser generation bands (along with a certainly strong difference in gain) in frames of the theory[2] signifies a distinction in wave coupling constant in these spectral regions. Considering that wave coupling constant of CLC-based DF-laser is proportional to LC birefringence, data obtained testify that these two generation bands are caused by periodic structures with differing contribution

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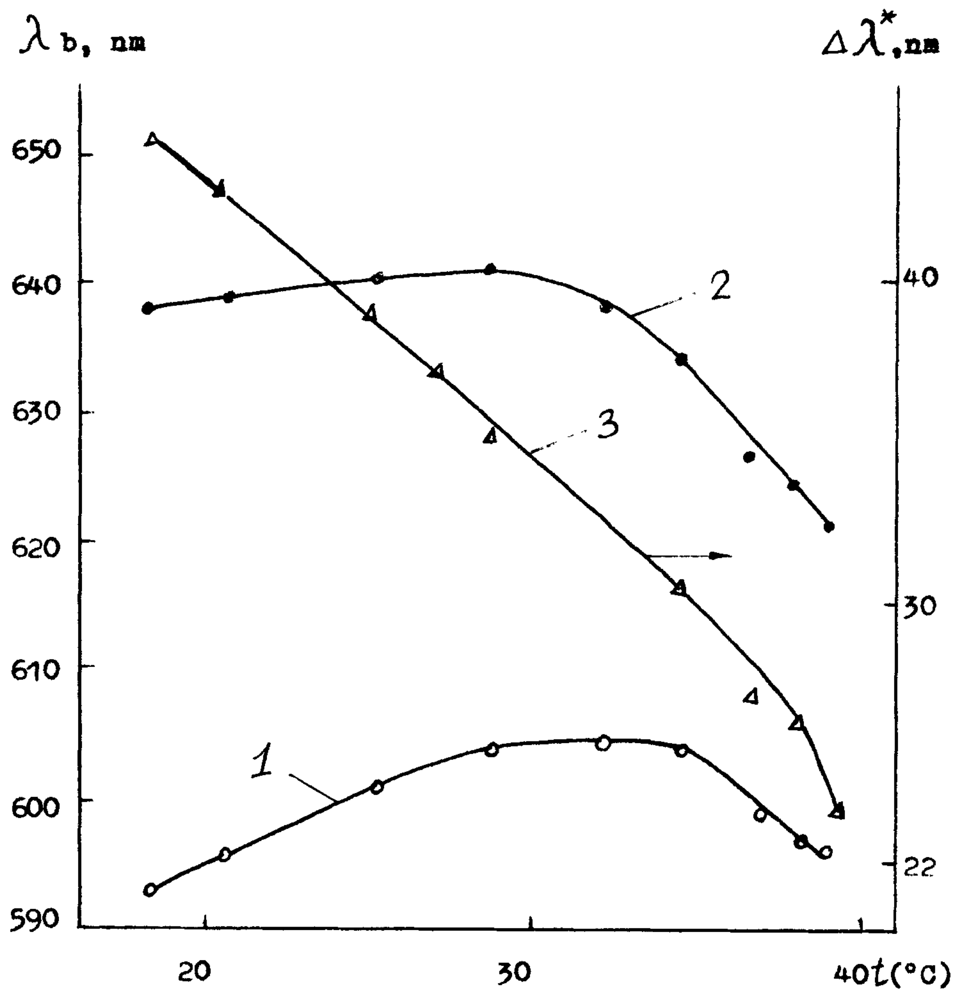

FIGURE 2 Temperature behaviour of the wavelength of central line in short- (1) and long-wavelength (2) DF-laser generation bands (sample II). 3 - temperature change of the distance between both bands (Δλ*).

to birefringence for the light passing along a helical axis.

Linear polarizer and $\lambda/4$-plate were used for a determination of generation band polarization. Left-hand circular polarization appeared to coincide for both generation bands with the sign of helicity of induced CLC.

Temperature dependences of a triplet's central line wavelength of these bands for the sample II are shown in Fig.2. As it could be seen, their temperature behaviour points to the inversion of the sign of helical pitch change at the temperature about 30 ^{o}C. It is also verified by the temperature change of λ_B of selective reflection band (Fig.3). However, the rate of this shift essentially differs for short- and long-wavelength generation band (Fig.2) under as well as above the inversion point. For the short-wavelength band (curve 1, Fig.2) $d\lambda/dT$ is 1.1 nm/deg under and -2.5 nm/deg above the inversion temperature, for the long-wavelength band (curve 2, Fig.2) it is 0.35 nm/deg and -3.3 nm/deg respectively. Spectral distance ($\Delta\lambda^*$) between generation bands, shown in Fig.2 (curve 3), as well as selective reflection halfwidth (Fig.3, curve 2) apparently linearly decrease while the temperature rises. For the selective reflection band such variation of $\Delta\lambda$ with the rate of -2.25 nm/deg is trivial and caused by the temperature change of birefringence. The rate of the change for $\Delta\lambda^*$ is -0.9 nm/deg and its difference from that for $\Delta\lambda$ points to the influence of additional reasons as, for example, different temperature dependences of elastic constants.

The experimental data presented here may be interpreted as a consequence of the harmonic distortion of the helical structure of induced CLC. As it follows from the ratio of the threshold excitation intensity to the fluorescence intensity on the generation wavelength for the either of the bands for the sample II, wave coupling constant and "resonator efficiency"[9] of DF-laser is greater

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for a long-wavelength band. It might be associated only with the larger contribution of a periodic structure with the pitch corresponding to the long-wavelength band to birefringence.

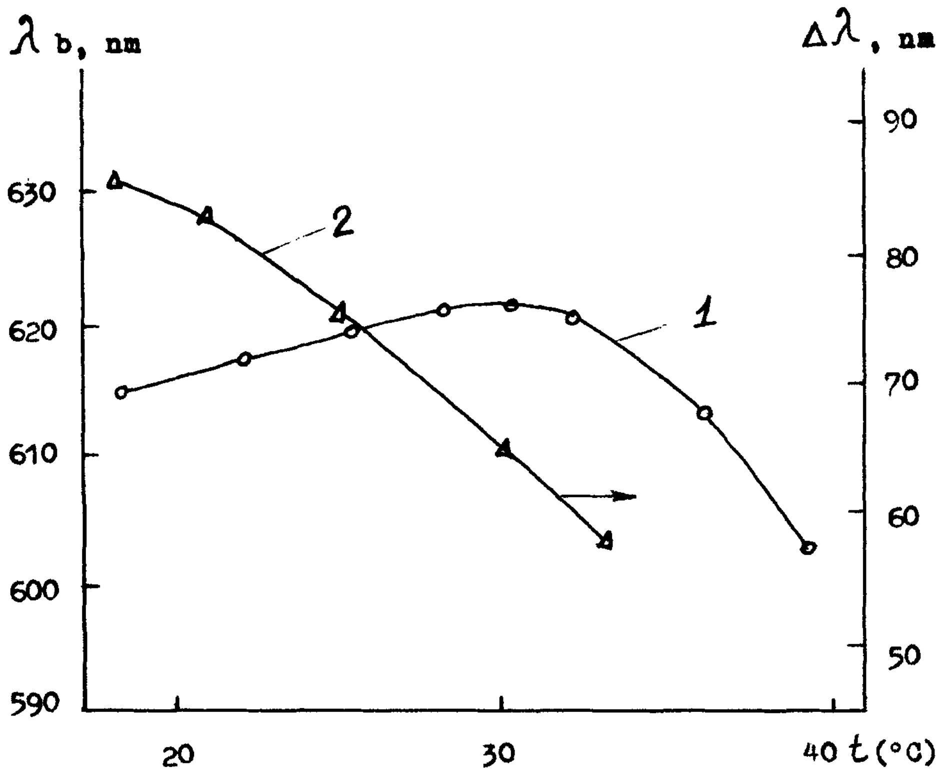

FIGURE 3 Temperature behaviour of the wavelength λ_B of SR-band centre (1) and halfwidth (2) for the sample II.

Thus, we consider this generation band as corresponding to the period of a helical twisting itself and being caused by Bragg scattering of a gained light in quasi-nematic layers parallel to cell substrates. The equality of the threshold excitation intensity of the generation bands for the sample II, despite the essentially greater fluorescence intensity in the spectral region of the short-wavelength band

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(Fig.1), points to the relatively lower "resonator efficiency" and, thus, comparatively smaller contribution of the periodic structure corresponding to this band to the birefringence value. We suggest that the short-wavelength generation band is caused by the period of static solitons, which are the alternating regions of LC-director deflection from quasi-nematic layers' plane at the certain angle θ.[6,8]

The data of polarization measurements allow to ascertain a spatial arrangement of static solitons with reference to the helical axis. Their linear arrangement along the axis will result in the linearly polarized generation of the appropriate band as the fluorescence radiation of both handnesses will scatter in the same way. The dopant's fluorescence radiation only of the circular polarization corresponding to the sign of helix is known[1] to scatter in the helical periodic structure. The registered circular polarization of both generation bands, with the same handness as that of the helix, corroborates the helical arrangement of static solitons.

Divergence in temperature dependences of the generation bands testifies the statement about their differing physical nature resulted from threshold excitation intensities. As it could be seen if comparing Fig.2 and Fig.3, temperature behaviour of the wavelength of the centre of SR band λ_B much better corresponds to that of long-wavelength generation band. However, strict agreement of their rates ($d\lambda/dT$) is not observed possibly because of the experimental inaccuracy. Strong difference in temperature-stimulated changes of the halfwidth of SR band $\Delta\lambda_B$ and generation interband distance $\Delta\lambda^*$ also points out different physical nature of the bands. It should be noted that there is another possible reason for the existence of two-band generation in induced CLC. If the conditions of Kogelnik theory[2] are not valid for the real CLC, strong dying down of diffracting wave in the centre of SR-band results in generation of two symmetric bands at the edges

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of SR-band. There is no selectivity in this case and a feedback would occur due to reflection at the CLC/substrate boundaries. Weak efficiency of such resonator would result in a great increase of generation threshold. In particular, Fresnel-reflection-caused generation was not observed in DF-lasers based on steroid CLC over the whole pumping intensity range. As the birefringence of the studied CLC compound is $\Delta n=0.15$ and average refractive index is $n \cong 1.5$ the main condition of the theory[2] is valid ($\Delta n << n$) for the phase grating in it as well as for steroid CLC ($\Delta n=0.05$ and $n=1.5$). Moreover, the contribution of the additional thermal phase grating, arising under the strong excitation due to the partial generating dye alignment, is negative (according to[9]) and diminishes the real Δn of initial periodic structure.

Formation of the second spatial period in the CLC structure could not be attributed to the doping of the laser dye as its concentration is low (0.3 wt.%) and its twisting activity is much lower than that of steroid CLC (36 wt.%). Besides that, we used the same dye as in the DF-laser on the mixture of steroid CLCs, where no additional generation bands were observed. The form and spectral position of absorption band of the dye in steroid and induced CLC as well as in nematic MBBA mesophase testify that dye molecules do not aggregate and do not form complexes with LC's molecules. It is corroborated also by the coincidence of the shape and intensity of the dye's fluorescence spectrum in CLC and in alcohol solution (quantum efficiency is nearly 50% under the equal dye's concentration). So, the solution of the dye in the induced CLC as well as in steroid one could be considered to be the molecular one.

On the basis of the experimental results presented, we could make a conclusion that harmonic distortions of the helical structure of the period smaller then that of the own helical pitch are observed in studied mixtures of CLC and NLC.

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