Generation of a tunable radiation by impurity cholesteric liquid crystals

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A generation of light in dye-activated oriented layers of cholesteric liquid crystals (CLC) was obtained for the first time. The spiral structure of the CLC provided a distributed feedback. Temperature tuning of a narrow generation band was obtained experimentally as a result of temperature variation of the pitch of the cholesteric spiral.

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The spiral twist of CLC molecules in an oriented planar texture gives rise to a phase grating through the thickness of the layer with a period equal to one-half the spiral pitch P. In such a medium for normal light incidence the radiation with a circular polarization with the same sign as the spiral twist is totally reflected in the first order,[1] and the Bragg condition has the form $P\bar{n} = \lambda$, where λ is the wavelength of selective reflection and $\bar{n}$ is the average refractive index of the CLC.

The selective properties of the CLC and the ability to change the spiral pitch[2] are of interest for building frequency-tunable lasers. It is important that it is possible to combine the major functional elements of a laser—active medium, cavity, and tunable selector—in a film. The idea of building a laser with distributed feedback using a CLC and some theoretical estimates of its operation were given earlier.[3,4]

In this paper we report the first construction of such a laser, which combines the selective reflection band of the CLC matrix with the maximum gain region of the dye dissolved in it.

A three-component mixture was used as the CLC: cholesteryl chloride + cholesteryl oleate + cholesteryl pelargonate, similar to that investigated earlier[5] and present in the cholesteric mesophase at room temperature. The spiral pitch in such a mixture varies linearly with the temperature $(dP/dt > 0)$.

The planar texture was formed by filling a cell consisting of two plane-parallel plates of fused quartz separated by 40 to 250-μm-thick gaskets, and the mixture had a dye dissolved in it. The inner walls of the plates, coated with a transparent SnO_2 film, were rubbed in one direction to orient the CLC boundary layers. To improve the quality of the planar texture for cell thicknesses $> 80\ \mu$m, it was subjected to repeated ageing, consisting of heating the mixture above the temperature for the phase transition to the isotropic state, followed by a slow cooling and a shifting of the plates in the rubbing direction at the phase-transition temperature.

Suitable dyes were obtained from the benzanthrone derivatives, which possess excellent solubility and a high quantum yield (40–50%) in the CLC.

Figure 1 shows the transmission (a) and fluorescence (b) spectra of the dye, an analog of benzanthrone in the CLC mixture. The circular dichroism absorbed by the dye, which was observed in the transmission spectra for the two circular polarizations, is attributable to the low ordering in the matrix, since the benzanthrones and their derivatives are not isomorphic impurities in the liquid crystal. The ordering of the imbedded dye, which was measured by us from the linear dichroism of the absorption
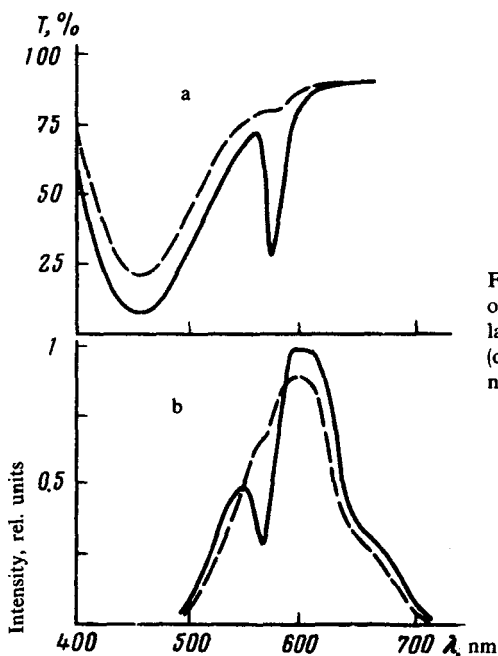

FIG. 1. Transmission (a) and fluorescence (b) spectra of the investigated dye in a CLC:——left-hand circular polarization,----right-hand circular polarization (dye concentration is $c = 3.2 \times 10^{-2}$ m/l, layer thickness is 40 μm, temperature is 20 °C.)

in the nematic MBBA, is equal to 0.22 (we did not know the exact value of n_0 and n_e for the CLC mixture).

The maximum reflectivity (575 nm) for the CLC layer with a dye is 5–6% lower than that for the pure CLC matrix in a 40-μm-thick layer. At the same time, the reflection maximum of a pure CLC layer of this thickness is displaced by 8 nm toward longer wavelengths. If the dye absorption in Fig. 1a at a concentration of $\approx 10^{-2}$ m/l is noticeable in the selective-reflection region, we can account for this shift by the Borman effect in absorbing CLC's.[6]

The selective properties of the matrix are also evident in the fluorescence spectra of the dye (Fig. 1b). The fluorescence in this case is characterized by certain unique features, which, so far as we know, have not been observed before. When the fluorescence occurs in the normal direction to the oriented layer (along the CLC spiral axis), the spectrum has a dip for the left-hand, circularly polarized radiation, which coincides in frequency with the selective reflection of the matrix (Fig. 1a). The deformation of the right-hand, circularly polarized fluorescence spectrum correlates with the transmission spectrum of this polarization (Fig. 1a). A deformation in the selective-reflection region indicates that the circularly polarized fluoresence radiation occurs only in this region, and the remaining regions of the fluorescence spectrum are elliptically polarized.

For studying the lasing, we pumped a pulsed dye laser excited by the second harmonic of a ruby laser ($\tau_p = 30$ nsec). The active medium of the dye laser was a solution of TOPOT in dioxane that lased in the 424 to 436-nm spectral interval. The dye laser with a power density of 200 kW/cm² in a 1.5-mm diameter spot without additional focusing was directed at the cell containing the dye solution in CLC, which was placed in a thermostat (the accuracy of the temperature stabilization was 0.1 °C). Lasing was done at the optical densities of the dye solution of 4–6 at the pumping
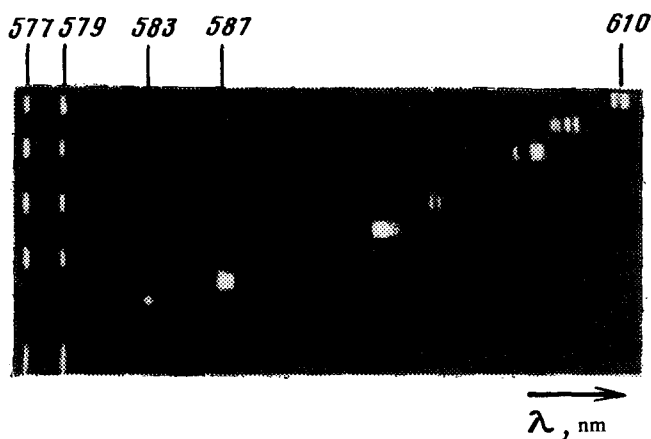

FIG. 2. Lasing spectra and lasing frequency tuning as a result of varying the temperature from 22.5 to 31.5 °C (layer thickness is 200 μm, dye concentration is $c = 6.4 \times 10^{-3}$ m/l).

frequency in the direction of the optical axis of the CLC. A reduction of the thickness of the CLC layer with the dye to $< 60\,\mu$m without changing the optical density of the solution increased the threshold and stopped the lasing because of concentration quenching of the dye fluorescence. The threshold power density of 200 to 250-μm-thick cells was 60 kW/cm^2 and the lasing bandwidth at the threshold was 0.3 nm; the band was broadened to 0.7 nm as a result of three-fold enhancement of the threshold.

Figure 2 shows the lasing spectra and temperature tuning of the lasing frequency. The radiation was tuned in the range 583–610 nm by heating the cell in the interval of 22.5 to 31.5 °C. The tuning rate was 3 nm/deg. The lasing spectra in Fig. 2 correspond to a rapid variation of the cell temperature (1 °C in 5–7 min). A high variation rate of cell temperature increases the lasing line width to 1–1.5 nm. A reduction of the CLC heating rate to 1 °C in 15–20 min was accompanied by a narrowing of the lasing line width to 0.3–0.5 nm. Such broadening is apparently attributable to the fact that the spiral pitch is different from the equilibrium pitch in the CLC because of its low thermal conductivity.

Our investigation, therefore, confirms the possibility of building small-size, thin-film tunable lasers employing CLC's. This opens up the prospects for building fundamentally new devices using such lasers. The frequency tuning rate of these lasers can be increased significantly by unwinding the CLC spiral with an electric field.

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