THE DISTRIBUTED FEEDBACK LASER BASED ON DOPED NEMATIC LIQUID CRYSTAL

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We have studied distributed feedback (DFB) lasers based on doped nematic liquid crystals (NLCs). Two optical schemes for the formation of an interference pump distribution are considered, namely, the excitation by a co- or counterpropagating pumping beam. The spectroscopic features of a number of dyes as NLC dopants are analyzed, and pyromethene dyes are adopted as the most promising for the DFB-laser fabrication. The conditions needed for a laser with dynamic DFB in NLC and a similar laser with static DFB in a cholesteric liquid crystal (CLC) to operate are compared. The modulation depth of a phase grating, which governed the lasing threshold, turned out approximately two orders of magnitude larger in CLCs than in NLCs, provided the dynamic formation of the grating by the laser pump field. The lasing by an NLC on the dynamic DFB has been obtained for the first time. Various ways to optimize the key parameters of the lasing with frequency tuning are analyzed and experimentally tested for both pumping schemes.

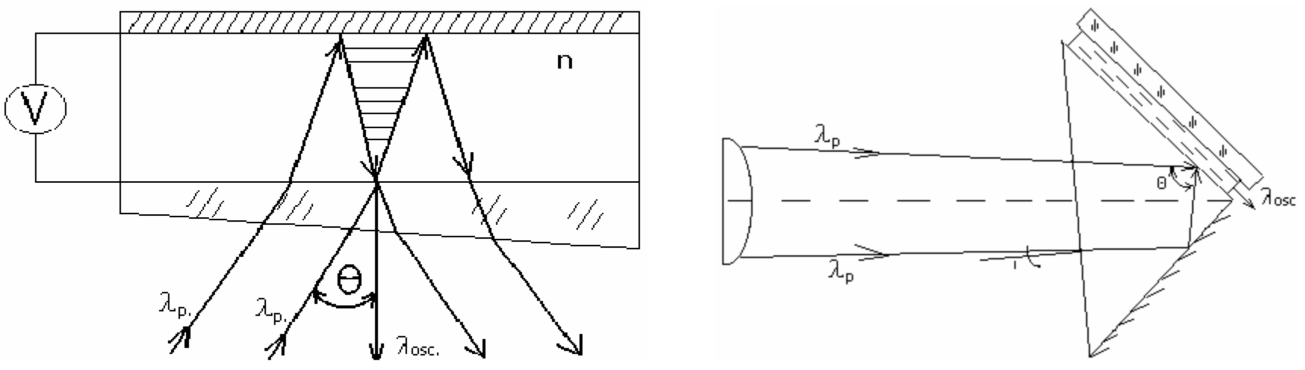
Liquid crystal lasers are actively studied today at a good many scientific centers throughout the world, because, besides the scientific interest, there exists a probable opportunity for their practical use, in particular, in the up-to-date information display facilities. For today, certified are NLC-based lasers with a mirror resonator [1, 2] and those of the wave-guide type in the superluminescence mode [3], as well as lasers with static DFB on the basis of CLCs [4–8]. A characteristic feature of a DFB laser of any type is the absence of a mirror resonator, since the positive feedback for the generated radiation is realized here due to the Bragg scattering by a periodic structure that either exists or is created in the active medium. CLC-based DFB-lasers [4] are especially attractive for the creation of information display systems with enhanced brightness, because the natural helical structure of CLCs, which ensures static DFB, allows the active medium of such lasers to be fabricated in the form of a screen with arbitrary area and curvature.

However, the problem of the effective and controllable lasing frequency tuning in such lasers remains challenging till now. An inertial temperature-induced variation of the lasing frequency, which is used in lasers of such a type today, cannot be considered promising. The application of an electric field along the axis of the CLC helix, provided that the CLC is characterized by a positive dielectric anisotropy ($\Delta\varepsilon > 0$), so that NLC molecules rotate under the field action, gives rise to a periodic deformation of the planar structure of CLC, with the helix axis being rotated by $\pi/2$ at the subsequent increasing of the field strength [9]. Such a deformation drastically worsens the optical transparency of the liquid crystal and makes the lasing impossible under those conditions. Another geometry of imposing the electric field, i.e. normally to the helix axis, is of little use for practical applications, first of all due to the necessity to apply high voltages of more than 1 kV.

In the case of lasers based on doped NLCs, the solutions of this problem can be obtained by reorienting the director and, respectively, by changing the refractive index of the crystal under the action of an electric field [9]. However, such an electrically tunable DFB-laser based on NLCs activated by dyes has not been created yet. Therefore, the main task of this work was to study the conditions needed for a laser with dynamic DFB and frequency tuning by varying the induced grating period to be created.

In the experiments, we used two types of commercial-grade nematic liquid crystals, mixture LC-654 and cyanobiphenyl 5CB. These NLCs were activated by means of the introduction of laser dyes belonging to different classes. The concentration of dyes in the solution was 0.2–0.3 wt.%.

The specimen for studying the optical, spectral, and lasing properties was a cell composed of two glass substrates. The internal side of one of them was covered by an aluminum layer. The other, wedge-like substrate (the wedge angle was about 3°) remained transparent. To align the doped NLC, the internal side of each substrate was additionally coated with a polymer layer, polyimide varnish (PIV), less than 1 μm in thickness and rubbed in one direction. To create an aligned layer with a planar structure, the NLC was heated up to the isotropic state and, due to capillary forces, it was drawn into the gap between the substrates. The NLC layer thickness



a b

Fig. 1. Excitation schemes of a DFB-laser on the basis of NLC: (*a*) with a mirror cell and (*b*) with a TIR prism

in this design was ensured making use of fluoroplastic spacers 50–300 μm in thickness.

In another design, the aligned layer of doped NLC with planar structure was formed between a lateral face of the total reflection prism and the plane-parallel glass substrate; the both were preliminary covered with a rubbed PIV layer. In this specimen, the layer of the aligned liquid crystal was 50 μm in thickness.

The transmission spectra of doped NLC specimens were registered on an SF-20 spectrophotometer. The specimens were excited by the pulse ($\tau_i \approx 20$ ns) emission of the second harmonic of a Nd^{3+}-laser ($\lambda = 530$ nm). This laser operated in the mode of passive Q-factor modulation, and the repetition frequency was 1 pulse/min. The energy of pumping pulses was controlled by neutral optical filters and monitored making use of a calorimeter. The lasing and superfluorescence spectra of doped NLCs were registered directly, by means of a camera-recorder, from a ground glass with a graduated wavelength scale located in the cassette section of the spectrograph with the inverse dispersion of 0.6 nm/mm.

The concept of dynamic DFB-laser on the basis of doped NLC with electric frequency tuning involves an instant record of a spatially periodic pattern of the dye population by the pumping field. A simultaneous imposition of the electric field, for the sake of changing the director orientation and, accordingly, the refractive index of NLC for a certain polarization of the incident light, gives rise to a variation of the lasing frequency.

To build a dynamic DFB-laser, we used the following excitation schemes. Figure 1,*a* exhibits a scheme, in which the spatial periodicity in the doped NLC layer is induced due to the interference between two pumping beams, the incident one and the beam reflected from the mirror substrate. Provided that a plane-aligned NLC with a positive dielectric anisotropy is used in such a cell, the imposition of an electric field can change the inclination angle of the optical axis and, accordingly, the refractive index of the liquid crystal, which gives rise to a variation of the lasing frequency.

The lasing wavelength λ_{osc} in the given excitation scheme of a DFB-laser based on a liquid crystal depends on the incidence angle θ of exciting beams and, provided that the Bragg condition is fulfilled exactly, it is determined by the expression

$$\lambda_{\mathrm{osc}} = \lambda_p n/(n^2 - \sin^2\theta)^{1/2}, \tag{1}$$

where λ_p is the excitation wave length, and n the refractive index of the liquid crystal.

The excitation scheme with a total internal reflection (TIR) prism (Fig. 1,*b*) is widely used for dynamic DFB-lasers based on isotropic solutions [10]. In this case, the lasing wavelength is determined as follows:

$$\lambda_{\mathrm{osc}} = \frac{n\lambda_p}{n_{\mathrm{pr}}\sin\theta} = \frac{\sqrt{2}n\lambda_p}{\sqrt{n_{\mathrm{pr}}^2 - \sin^2 i} + \sin i}, \tag{2}$$

where n and n_{pr} are the refractive indices of NLC and a prism, respectively; and i is the incidence angle of the exciting irradiation onto the diagonal face of the prism.

According to work [11], the threshold gain coefficient α of a DFB-laser at the frequency ω is determined by the expression

$$\exp(2\alpha L) = \frac{4}{M^2}\left[\alpha^2 + \left(\frac{n\Delta\omega}{c}\right)^2\right], \tag{3}$$

where $M = \pi n_1/\lambda + i\alpha_1/2$ is the coupling constant (the coupling arises owing to the Bragg scattering by the phase and amplitude gratings with modulation amplitudes n_1 and α_1, respectively), $\Delta\omega$ is the frequency detuning from the exact Bragg frequency, and L is the active region length.

Relative contributions from the phase and amplitude gratings to the lasing threshold for lasers with dynamic DFB, based on dye solutions, were estimated in work [12]. Let us make a similar estimation for a DFB-laser based on NLC, taking into account that the coefficients of refractive index non-linearity of alcohols and NLCs are close to one another [13]. The refractive index is known to change in the strong light wave field by engaging a number of mechanisms: the Kerr effect, electrostriction, and thermal heating of the substance:

$$n = n_0 + n_2 I, \tag{4}$$

where n_0 is the refractive index in the absence of a strong light wave, n_2 the coefficient of refractive index non-linearity of the active medium, I the intensity of a light wave, and $n_2 I$ a nonlinear increment to the refractive index caused by the powerful light wave. At pumping powers typical of the laser generation, the nonlinear increment $n_2 I$ given by electrostriction and the Kerr effect is about 10^{-9} and 10^{-10}, respectively, for alcohol solutions [13].

If, owing to the laser excitation, light is absorbed directly, a change of the refractive index of either alcohol or NLC can be determined as

$$\Delta n_t = (dn/dT)_p \Delta T, \tag{5}$$

where ΔT is the temperature increment from the equilibrium one, and $(dn/dT)_p$ is the temperature dependence of the refractive index at a constant pressure, when the pump pulse duration exceeds the sound propagation time in the excited region of NLC.

In the absence of the heat transfer (for nanosecond time intervals), the temperature variation is equal to $\Delta T = Q/(V\rho C_p)$, where Q is the amount of heat that is released in the NLC irradiation region, C_p the specific heat at a constant pressure, V the pumped region volume, and ρ the density of a solution. To estimate the amount of heat that is released in the active region of a solution, we use the relation $Q = E_n(1-\Sigma)\lambda_n/\lambda_g$, where E_n is the pump pulse energy, Σ the energy efficiency of a dye laser, and λ_n and λ_g are the pumping and lasing wavelengths, respectively. In the case of typical spectroscopic parameters of rhodamine 6G dye in an alcohol solution and an excitation energy of a few millijoules, we obtain that $\Delta n_t \approx 10^{-4}$.

The estimations made for isotropic solutions remain valid for NLCs at their excitation by nanosecond pulses, when non-linearities provoked by the Kerr effect and electrostriction are 4 to 5 orders of magnitude lower than the thermal one [14]. These estimations demonstrate that the threshold conditions for a DFB-laser on the basis of doped NLC, which depend on the coupling coefficient M, are much worse than those for a DFB-laser based on doped CLC, provided equal thicknesses of the active region L. It is so, because the modulation depth of the refractive index that arises owing to the nonlinear interaction with the pumping field is two orders of magnitude smaller in the NLC case than the modulation depth in the natural helical structure of CLCs. Therefore, in order to achieve the threshold lasing conditions in the scheme exhibited in Fig. 1,*a*, it is necessary to increase the thickness of the active layer as much as possible and to create a high-contrast interference pumping field.

To create a DFB-laser on the basis of NLC, dyes of various types were used: neutral benzanthronic and phenolenone ones, as well as ionic polymethine ones with absorption in the green spectral range. The indicated dyes were characterized by a rather low quantum yield of fluorescence in NLCs (less than 10%), so that the threshold lasing conditions were not reached in them at thicknesses up to 250 μm even at maximal—limited by a destruction threshold—intensities of excitation [15]. Among pyromethene dyes, we found those that manifested suitable spectroscopic characteristics in the green+yellow spectral range for the lasing in NLCs at the nanosecond excitation. These dyes are characterized by a good solubility and a high quantum yield of fluorescence in NLCs. The solution of pyromethene dye No. 567 in LC-654 with an absorption maximum at 524 nm and a fluorescence maximum at 548 nm has a quantum yield of fluorescence of 98%.

In Fig. 2, the absorption spectra of this dye in NLC LC-654 are shown for two linear polarizations of light. One can see that the absorption dichroism of the given dye in NLC is not enough to substantially affect the lasing threshold on the reorientation of the liquid crystal by an electric field. The laser generation in NLC doped with this dye was obtained using the excitation scheme depicted in Fig. 1,*a*. The wedge-like layer of doped NLCs with an average thickness of 250 μm was used. The lasing threshold was comparable with that for a layer of R6G alcohol solution with the same thickness.

In Fig. 3, the lasing spectrum of doped NLC is presented. It was registered with the help of a camera-recorder on the surface of a ground plate in the spectrograph. The incidence angle of the exciting beam was 35°. The average lasing wavelength was 554 nm,

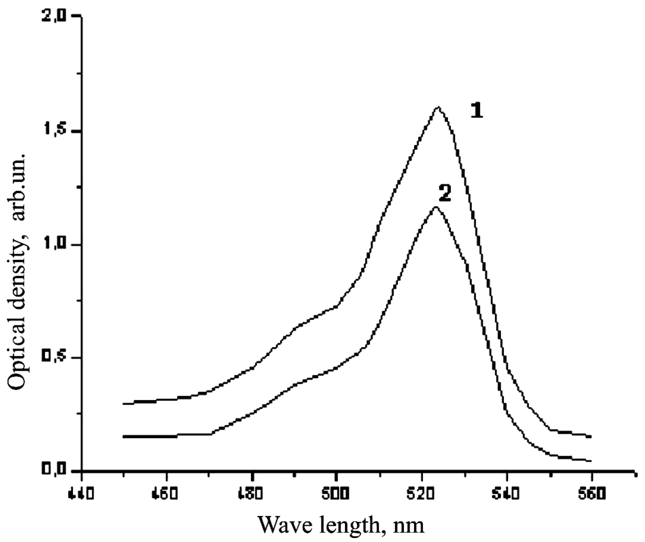
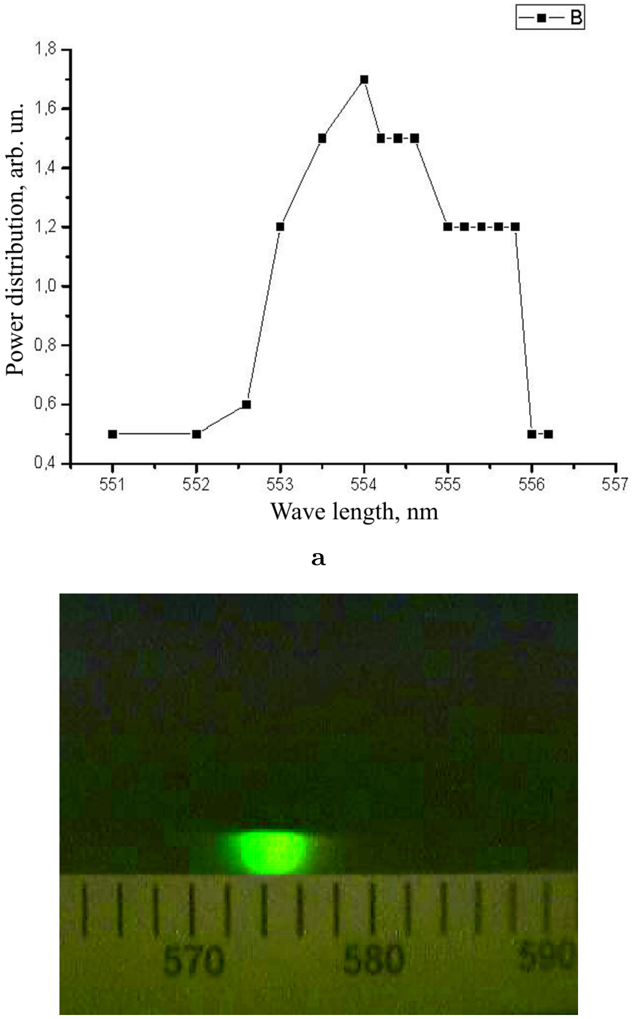


Fig. 2. Optical absorption dichroism in pyromethene dye No. 567: absorption for a linear polarization of light along (*1*) and normally (*2*) to the NLC director



a

b

Fig. 3. (*a*) Lasing spectrum of NLC LC-654 activated by pyromethene dye No. 567 in the excitation scheme depicted in Fig. 1,*a* (b) superfluorescence spectrum of NLC 5CB activated by pyromethene dye No. 597 in the excitation scheme depicted in Fig. 1,*b*

and the spectrum width was about 1.45 nm. A wider diffusion pedestal 4 nm in width was caused by a superluminescence contribution. The researches of the lasing spectrum at various excitation angles showed that the spectral width changes in such a manner that the narrow line arises only in a definite range of excitation angle (28 to 30°).

Our researches of the conditions needed for a DFB-laser based on doped NLC in a cell with a mirror substrate to lase demonstrated that such a scheme is favorable for the emergence of superfluorescence. This phenomenon stems from the light reflection in the mirror–wedge-like substrate system, in spite of the presence of a 3°-wedge in both the active substance and the substrate. To exclude superfluorescence, we used the scheme with a total reflection prism (Fig. 1,*b*). A layer of liquid crystal 5CB doped with pyromethene dye No. 597, which is characterized by absorption in a longer-wave range, was placed between the prism and the aligning transparent substrate. In experiment, we used the geometry, when the refractive index of the liquid crystal for the vertically polarized pumping wave equaled n_0. In the case of pyromethene dye No. 597 solution in 5CB, we observed the intensive superfluorescence with a maximum at 575 nm and a pedestal about 4 nm in width at the incidence angle $i = 11°$ (Fig. 3,*b*). The results of calculations of the average wavelength of this spectrum for an incidence angle of the pumping wave of 11° gave good agreement with the experimentally observed band. Note that we have not yet obtained a narrow lasing line using this scheme, because we have not optimized the active layer thickness to minimize the influence of the waveguide effect on the lasing spectrum.

The results obtained till now allow us to draw the following conclusions:

1. A narrow lasing line, which is inherent to a DFB-laser on the basis of NLC and obtained in the scheme with a mirror cell, is accompanied by a broadband superfluorescence with a low excitation threshold, which

arises due to the light reflection from cell's surfaces. One of the ways to increase the ratio between the power of the main lasing line and the superfluorescence background is to reduce the transverse cross-section of the exciting radiation.

2. To achieve a low lasing threshold for DFB-lasers on the basis of NLCs, it is necessary to use solutions with the optical density of not above 3 and with the thickness of the aligned NLC layer as large as possible, which agrees with with theoretical formula (3) derived by A. Yariv [11].

3. The pumping scheme with a total reflection prism excludes the opportunity of a feedback, which could arise due to the light reflection from the cell's surfaces. This scheme is promising from the viewpoint of the complete exclusion of superfluorescence by the optimal choice of the active region length L, whose value determines a chaotic feedback due to the nonresonant inverse scattering in the NLC layer.

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ЛАЗЕР З РОЗПОДІЛЕНИМ ЗВОРОТНИМ ЗВ'ЯЗКОМ НА ДОМІШКОВОМУ НЕМАТИЧНОМУ РІДКОМУ КРИСТАЛІ

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Резюме

Досліджено лазери з динамічним розподіленим зворотним зв'язком (РЗЗ) на основі домішкових нематичних рідких кристалів (НРК) при двох схемах формування інтерференційного розподілу накачки: збудження на зустрічних та попутніх пучках накачки. Вивчено особливості ряду барвників в ролі лазерних домішок в НРК і показано, що перспективнішими для створення РЗЗ-лазерів є пірометенові барвники. Порівняно умови реалізації лазера з динамічним РЗЗ в НРК з аналогічним лазером зі статичним РЗЗ на холестеричних рідких кристалах (ХРК). Для останніх глибина модуляції фазової ґратки, що визначає поріг генерації, приблизно на два порядки більша, ніж в НРК в умовах динамічного формування ґратки полем лазерної накачки. Лазерну генерацію НРК при динамічному розподіленому зворотному зв'язку отримано вперше. Проаналізовано і апробовано шляхи оптимізації основних характеристик генерації зі зміною частоти для вибраних схем збудження.