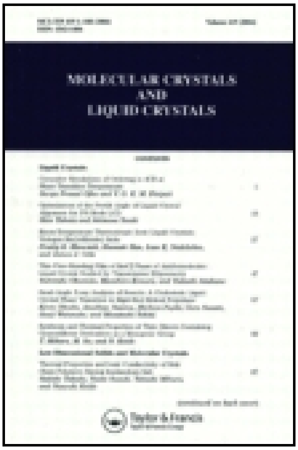

Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl20

Lasing in Dye-Doped Nematic Liquid Crystals at a Dynamic Distributed Feedback for Two-Scheme Excitation

I. P. Ilchishin [a] , P. Yu. Maslov [a] , E. A. Tikhonov [a] , S. O. Lipnitsky [a] & A. A. Stepanov [a]
[a] Institute of Physics of the NAS of Ukraine, Kyiv, Ukraine

Version of record first published: 22 Sep 2010

Mol. Cryst. Liq. Cryst., Vol. 467, pp. 235–245, 2007

ISSN: 1542-1406 print/1563-5287 online
DOI: 10.1080/15421400701224074

Lasing in Dye-Doped Nematic Liquid Crystals at a Dynamic Distributed Feedback for Two-Scheme Excitation

I. P. Ilchishin
P. Yu. Maslov
E. A. Tikhonov
S. O. Lipnitsky
A. A. Stepanov
Institute of Physics of the NAS of Ukraine, Kyiv, Ukraine

The lasing of a dye doped nematic liquid crystal (NLC) under the dynamic distributed feedback (DFB) recorded in NLC for two-scheme pumping is considered. Estimations of the contribution of the amplitude and phase gratings to the forward-backward wave interaction in the arising periodic structure are made. The narrow frequency tunable emission against the background wide spectrum of the superluminescence is gained, by manifesting the oscillation under DFB. The spectral characteristics of some dyes in NLC important for lasing has been studied and tested under laser pumping. The obtained experimental results give a positive credit to the selected approach.

Keywords: distributed feedback; dye; frequency tuning; lasing; nematic LC

INTRODUCTION

A natural helix structure of cholesteric liquid crystals (CLC) doped by lasing dyes can be used to create compact optically excited lasers with distributed feedback (DFB) [1]. DFB in a CLC planar oriented texture is caused by the Bragg's scattering of a circular polarized light (the same sign as that of the helix structure) at the amplitude-phase helix gratings formed by the structure of CLC and absorption (gain) of a partially ordered dye in the CLC host. In order to gain lasing, the

We acknowledge our colleagues L.A. Dolgov and R.M. Kravchuk for making the NLC oriented layers on a TIR-prism and a mirror cell used in experiments.
Address corresponding to I. P. Ilchishin, Institute of Physics of the NAS of Ukraine, 46 Nauky Pr, Kyiv 03028, Ukraine. E-mail: lclas@iop.kiev.ua

overlapping of a pumping emission with the dye absorption spectrum and the dye fluorescence spectrum with the Bragg selective reflection (SR) spectrum of CLC should be provided. So the helix pitch determines the lasing wavelength.

The lasing of the dye-doped CLC attracts a considerable attention because of the unique possibility to develop a DFB laser with flexible non-mirror resonator of all cross sizes and curvatures.

Evidently, a very compact DFB laser can be created on this basis. Miniature light-emitted devices are of big interest for LC display technologies. For instance, they are suggested to multiply enhance the brightness of color projection screens [2]. The intense studies of such lasers based on new CLC-materials, being more technological than viscous cholesterol derivatives, are under way now [3–5].

The lasing frequency of CLC-based DFB lasers can be tuned by several methods. First, it can be controlled thermally. This method has been realized in the early experiments on CLC-based DFB lasers [1]. In those experiments, steroidal CLC was utilized with its typical rather strong temperature dependence of a helix pitch. The temperature dependence of the helix pitch was monotonous and fluent. However, the inertia of the temperature control, as well as the need of thermal stabilization of the laser medium, makes this method of tuning cumbersome and inefficient for practical applications. The other less sluggish ways of the helix pitch control, the stretching and compression of oriented layers [5,6], look also problematic for practical applications, because of the evident technical difficulties.

The actual task for the similar lasers is the research and development of new faster methods of the frequency tuning. As known, the electric field imposed along the axis of CLC spirals brings about their destruction and contributes to the strong scattering of light in such a material, which deteriorates the lasing conditions. In nematic liquid crystal (NLC), in which no natural spiral periodic structure exists, the optical axis of a liquid crystal can be reoriented by the electric field imposition.

In the present work, we propose a new method of the frequency tuning in DFB-lasers on NLC by the imposition of a DC electric field. The idea consists in the creation of a DFB-laser, whose spatial periodicity of gain/index refraction is resulted due to the interference of two crossed pumping beams. For our aim, NLC with the positive anisotropy of dielectric permeability is applicable. One can start from the planar orientation of such NLC and impose a DC field to make the smooth transition to the homeotropic state of orientation or vice versa. Correspondently, the average index refraction along the laser wave propagation $\langle n \rangle$ will change, and the phase lasing condition

$\lambda_g \approx 2\Lambda(\theta)\langle n\rangle$ will do the same (here, $\Lambda(\theta)$ is the spatial grating period). As a result, the lasing frequency tuning will take place.

EXPERIMENTAL

The lasing characteristics of a DFB-laser were studied on the experimental setup presented in Figure 1. A DFB laser was pumped by the second harmonic of a Nd^{3+} laser (530 nm) with passive Q-switch oscillating with a pulse duration $\approx$20 ns. The excitation energy was measured by a calorimeter IMO-2 and attenuated by a set of neutral light filters. The lasing spectra were registered by a diffraction spectrograph with an inverse dispersion of 0.6 nm/mm.

The planar texture layers of the dye-doped NLC were formed between glass substrates covered by rubbed polyvinyl alcohol aligning layers. The thickness of a NLC layer was maintained by teflon films with a thickness from 32 to 400 μm. The transmission spectra of such samples were measured by a spectrophotometer SF-20.

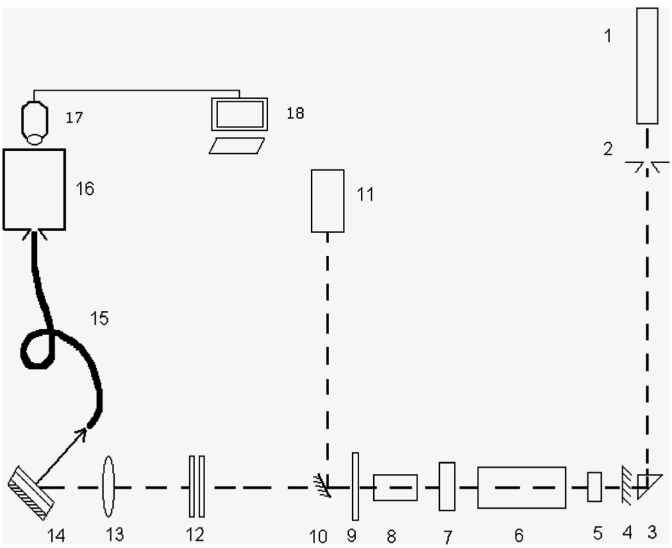

FIGURE 1 Experimental setup. 1 – He–Ne laser, 2 – stop's screen, 3 – reflecting prism, 4, 7 – mirrors, 5 – passive Q-switch modulator, 6 – active medium of a solid-state laser, 8 – nonlinear crystal, 9 – selective filter, 10 – dichroic mirror, 11 – calorimeter, 12 – neutral filters, 13 – lens, 14 – cell with active medium, 15 – optic-fiber light guide, 16 – spectrograph, 17 – web-camera, 18 – personal computer.

MOTIVATION OF THE METHOD

By analogy with the DFB lasing of isotropic dye solutions, we considered two optical pumping schemes for the case of anisotropic dyed NLC. For the first optical scheme (Fig. 2a), the lasing wavelength (λ_g) of the DFB depends on the incident angle of pumping beams and the refractive index of the active medium as follows:

$$\lambda_g = \lambda_{ex.} n/(n^2 - \sin^2\theta)^{1/2}. \qquad (1)$$

For the optical scheme depictured in Figure 2b, we have

$$\lambda_g = \frac{n_s \cdot \lambda_{ex}}{n_{pr} \cdot \sin\Theta} = \frac{\sqrt{2} n_s \lambda_{ex}}{\sqrt{n_{pr}^2 - \sin^2 i} \pm \sin i} \qquad (2)$$

Here, λ_{ex} is the excitation radiation wavelength, n_s is the refractive index of dye-doped NLC, n_{pr} is the refractive index of a prism, θ is

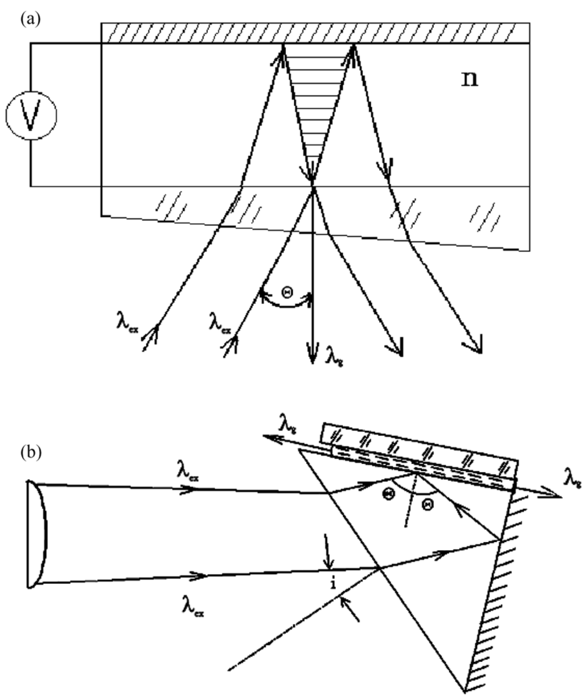

FIGURE 2 Two-scheme excitation in the DFB laser based on a dye-doped NLC: the scheme with the amplitude splitting of a pumping beam (a), the scheme with the wave front splitting of a pumping beam (b).

the incident angle of pumping beams in the active medium, i is the incident angle of beams on a prism.

In Figure 3, we present the calculated lasing wavelength of the proposed DFB-laser as a function of the refractive index for some different (double) incident angles in the scheme given in Figure 2. It is seen that a change $\Delta \boldsymbol{n} = 0.2$ (double refraction) results in the wavelength tuning of the DFB laser within 50 nm. The value is comparable with the dye fluorescence spectrum width.

To validate the nature of the proposed frequency tuning method, let us make some simplified estimations of the amplitude(gain)-phase grating which is responsible for the threshold oscillation in the DFB lasing. According to the theory of DFB-lasers on isotropic materials [7], the interaction power of counter running waves in a periodical structure is defined by the product of χ by the structure length L. In the gain-phase grating, both gratings will contribute to χ :

$$\chi = \pi \Delta n/\lambda + i\Delta\alpha/2, \tag{3}$$

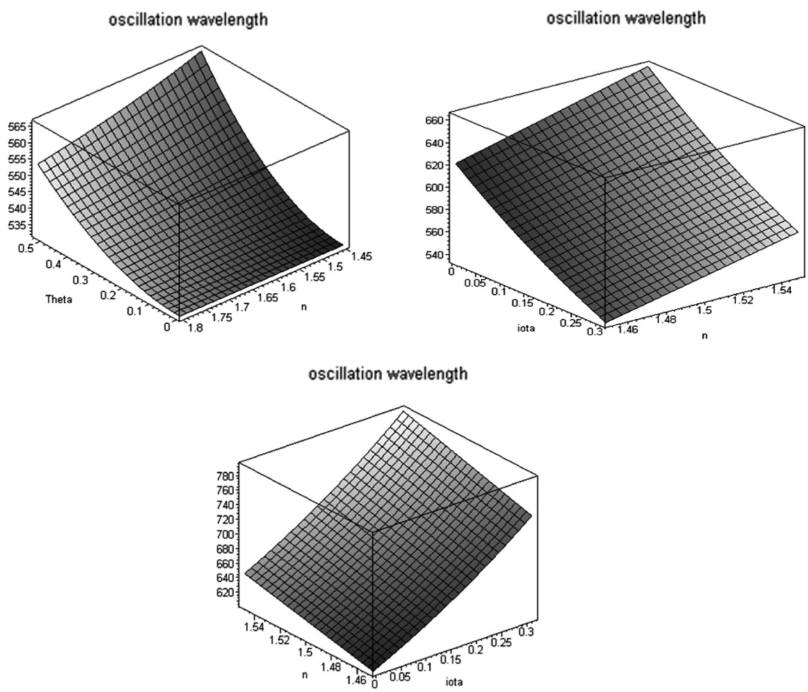

FIGURE 3 3D-dependences of the lasing wavelength on the refractive index and the incident angle of a pumping beam (in radians) in schemes (a) and (b) in Figure 2, respectively. In scheme (b), the prism index refraction $n = 1.8$.

where Δn and $\Delta\alpha$ are the index refraction and the gain modulation due to the interference pumping field recording in the dyed laser medium. The dynamical grating recording of the interference pumping field is resulted from a nonlinear optical response of the considered laser medium. Because the most optimal is the nano(pico) second pulse excitation for the dye lasing, when the thermal optical damage of the laser medium is minimal, no molecular orientational contribution to the nonlinear response in nano- and picosecond time domains exists [8]. Without this contribution, the nonlinear optical response of NLC is similar to that of the usual organic solvent. But the thermal contribution in a nonlinear response can stay important if $\Delta n = \tau_{ex}(dn/dT)_p$ is applicable (when $\tau_{ex} \geq \Lambda/\nu_{sound}$, where ν_{sound} is the sound velocity in a solution).

For a simplified evaluation of comparative contributions of each of the gratings (nonlinearity) in our experiment, we rewrite (3) in the squared form:

$$|\chi|^2 = (\pi\Delta n/\lambda)^2 + \Delta\alpha^2/4 \tag{4}$$

In a typical experiment with a DFB laser on the base of a R6G ethanol solution [9], $\Delta\alpha$ equals $\approx 4-5\,\text{cm}^{-1}$ and $\Delta\alpha\,^2/4$ comes to $\approx 4-6\,\text{cm}^2$. To estimate the contribution of the thermal phase modulation to χ, we can use the known thermodynamical relations, when the inequality $\tau_{ex} \geq \Lambda/\nu_{sound}$ is valid:

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

$$\Delta T = Q/C_v V_\rho, \tag{6}$$

where ΔT is the temperature jump from heating in the active zone, Q is the thermal energy, V is the volume active zone, ρ is the solvent density, and C_v is the specific heat.

The estimation of the amount of heat standing out in the active zone of a solution was done from the relation [10]

$$Q = E_p(1-\eta)\lambda_p/\lambda_g, \tag{7}$$

where E_p is the energy of a pumping pulse, λ_p and λ_g are the wavelengths of pumping and lasing emission, η is the lasing efficiency. For the typical spectroscopic parameters of dye R6G in ethanol and an excitation energy of several mJ [9], we have got the value $\Delta n_t \approx 10^{-4}$ and $(\pi\Delta n/\lambda)^2 \approx 25\,\text{cm}^2$. The contribution to χ from the electrostriction and the Kerr effect proportional to E^2 is by several orders less, because the field strength E becomes low in strongly absorbing media. So the thermal phase grating brings a dominant contribution to Δn. However, the gain grating within the dye amplification band occurs to be very important by affecting the oscillation mode spectra.

For a DFB laser based on CLC, the threshold density power is about $10\,\mathrm{kW/cm^2}$, $\Delta n \sim 10^{-2}$, and the layer thickness is $50\,\mu\mathrm{m}$. A similar evaluation for a DFB laser based on NLC, $\Delta n \sim 10^{-4}$. Therefore, it is necessary to enlarge the laser layer thickness. The restriction on the layer thickness of NLC oriented by walls was about $400\,\mu\mathrm{m}$.

RESULTS AND DISCUSSION

The comparable contribution to the interaction of counter running waves that defines the laser threshold shows the preferred effect of the phase-gain grating formed primary by a pumping beam to χ. These estimations have shown also that the condition for the performance of a dynamic DFB NLC laser based on a pumping-induced grating is much worse as compared with those for a DFB laser based on CLC with a natural grating. In order to overcome them, it is necessary to enlarge the laser layer thickness considerably and to record a quite efficient grating by the interference of pumping beams.

We are aware with some previous experience about functioning a dynamic DFB laser on the optical scheme given in Figure 2 with a dyed polymer and liquid solutions [11–12]. To liquidate the stray lasing, the the authors of these works used simply a wedge-operated layer of the active medium. This approach cannot be used for a NLC DFB laser, because the wedge-oriented layer will discover the different instabilities of the optical axis on the imposition of an electric field. That is why the multiple lasing frequencies in the zone of excitation are possible, but a similar spectral broadening of the laser line is undesirable.

For comparison, we have accomplished the experimental studies of the lasing thresholds and spectral characteristics of some dyed isotropic solutions. We used the lasing optical scheme presented in Figure 2a. We used an R6G ethanol solution filling cells with a thickness from $250\,\mu\mathrm{m}$ to 1 mm. To exclude the stray lasing due to the wall cavity of a cell, we use a transparent sphenoid substrate with wedge angle $\approx 2^\circ$. In this way, we try to find out the optimum dye concentration for the DFB lasing.

The dependences of the threshold energy pumping on the optical density of an R6G solution for different thicknesses (L) of layers are shown in Figure 4 . As seen, there exists the optimum area of the optical density in the limits $3 \div 6$ for different thicknesses of layers. Moreover, the threshold of lasing is higher as compared with that of a 3-dimensional DFB laser on R6G. At a layer thickness of $400\,\mu\mathrm{m}$, we registered the lasing spectra dependence on the incident angle.

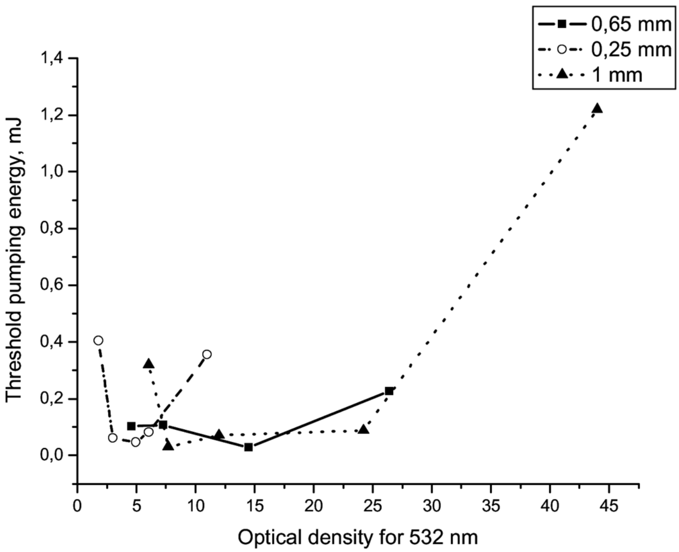

FIGURE 4 Dependence of the threshold lasing energy of the DFB laser based on an isotropic solution on the optical density for different thicknesses of the active medium.

However, we have not obtained the strong narrowing and the wavelength tuning which can be attributed to a real dynamic DFB laser.

In spite of the failure due to the low spatial coherence of a pumping beam (many-mode oscillation), the data on the optimal density of an R6G solution were used in the attempt to excite an NLC DFB laser. As a nematic liquid crystal, we used an industrial mixture ZhK-654 and 5CB. Both of them possess positive dielectric anisotropy. That is why, under the action of the field on the ensemble of molecules, its director will reorient along the field direction. For the dye doping of NLC, we used the different types of dyes: neutral benzantrone, phenolenone, and ionic polymethyne. All of these dyes discovered a very low quantum yield of fluorescence, and the lasing was not excited. We have found dyes with acceptable spectroscopic features for lasing in NLC under excitation by the second harmonic of an YAG-Nd laser amongst the class of pyrromethene dyes. They are characterized by a good solubility in NLC and a high quantum yield of fluorescence. The solution of such a dye in NLC with the absorption maximum at 524 nm and the maximum of fluorescence at 548 nm discovered the quantum yield of fluorescence of about 98%.

In Figure 5, we show the dichroism of the absorption band of this dye in a liquid crystal. The absorption of the dye is higher (curve 1)

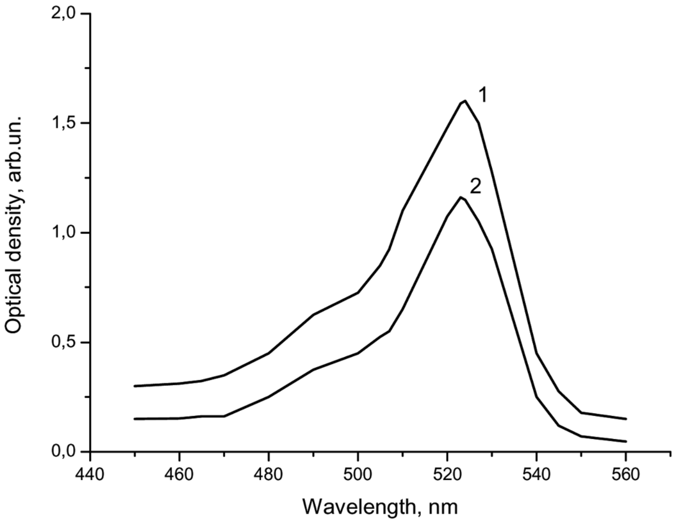

FIGURE 5 Absorption spectra of a pyrromethene dye (No 567) in NLC-654: 1 – light polarization is parallel to the director , 2 – light polarization is perpendicular to the director.

for the polarization of the incident radiation along the optical axis of the NLC crystal and decreases for the perpendicular polarization (curve 2). The dye lasing in NLC was obtained by the scheme presented in Figure 1. The layer of a dye-doped NLC in the form of a wedge was used, with an average thickness of 250 μm. The lasing threshold was the same as that for a solution of rodamine 6G.

Figure 6 presents the lasing spectrum of a pyrromethene-doped NLC registered by means of web-cameras directly with the opaque screen of a spectrograph with a scale of wavelengths under the angle of an incident pumping beam to be 35°. The average lasing wavelength is around 554 nm, and the total spectral width is close to 4 nm. As seen from the lasing spectrum presented in Figure 6, the narrow intensive

FIGURE 6 Lasing spectrum of a pyrromethene dye (No 567) in NLC Zhk-654.

line with a wavelength of 533.3 nm and the total width of 4 nm reveals itself against the background of a diffuse spectrum. The study of the lasing spectrum under the variation of an incident angle of the beam pumping has shown that the spectrum appearance is changed, and the narrow line appears only in a determined range of the excitation angles (28–30°). The study of the conditions of lasing in the DFB laser on a doped NLC in a cell with mirror substrate has shown that the stray oscillation easily arises in such a scheme, which is stipulated by the reflection from the mirror and transparent substrate, in spite of the presence of a 3°-wedge in the active medium and in the substrate. The possibility to separate the stray oscillation or even to suppress it relative to the DFB lasing in such a scheme is limited.

To completely suppress the stray oscillation, we adverted to another optical scheme which is presented in Figure 2b. In this scheme, a prism made of glass TF with the refraction index $n = 1.813$ is used. Its leg plane is silver-plated to have the full reflection under greater corners. The second leg plane is sheeted by an oriented layer, which is rubbed in one direction before assembling. The liquid crystal 5CB layer with the doping pyrromethene dye is placed between the prism and the transparent substrate. In our experiment, we realized the case where the refraction index of a liquid crystal for the vertical polarization of a pumping corresponds to n_o. The lasing wavelength in such a scheme of excitation of the DFB-laser is defined by formula (2). We have registered the intense superfluorescence band with a maximum at 575 nm and the width of about 4 nm, when turning the prism by an angle of 11°. The calculation of a lasing wavelength for an incident pumping angle of 11° relative to the prism gave a good correspondence with the experimentally observed superfluorescence band. So we met the problem of the strong broadening of the DBF lasing spectrum which should be overcome in the near future.

CONCLUSIONS

1. We have obtained the reliable operation of the DFB laser based on NLC in the scheme with a mirror cell hindering the stray broadband oscillation. One of the ways to overcome and to separate a narrow line from the broad spectrum consists in the use of a transparent substrate and the enhancement of the spatial coherence of the beam pumping.
2 We have shown that the lowest oscillation threshold of the DFB laser is achievable when the optical density of dye solutions is around 3 and the oriented NLC layer thickness is a few mm.

3 The optical scheme of a DFB laser with a prism is perspective if the stray oscillation is completely eliminated. However, it has great scattering losses in the NLC layer with a length of more than 10 mm as compared to the scheme with a mirror cell thickness of 250 μm. In addition, the scheme with a prism has technological difficulties concerning the creation of a uniform oriented NLC layer on the surfaces of a prism, which increases the dispersion and the lasing radiation line width.

REFERENCES

[1] Ilchishin, I. P., Tikhonov, E. A., Tishchenko, V. G., & Shpak, M. T. (1980). *JETP Lett.*, *32*, 24.
[2] Crawford, G. P., Firehammer, J. A., & Lawandy, N. M. (1998). *Liquid Crystals Today*, *8*, 7.
[3] Kopp, V. I., Fan, B., Vthana, H. K. M., & Genack, A. Z. (1998). *Opt. Lett.*, *23*, 1707.
[4] Taheri, B., Munoz, A. F., Palffy-Muhoray, P., & Twieg, R. (2001). *Mol. Cryst. Liq.-Cryst.*, *358*, 73.
[5] Finkelmann, H., Kim, S. T., Munoz, A., Palffy–Muhoray, P., & Taheri, B. (2001). *Adv. Mater.*, *17*, 1069.
[6] Umeton, C., Bartolino, R., Cipparone, G., & Simoni, F. (1988). *Appl. Opt.*, *27*, 210.
[7] Kogelnik, H. & Shenk, S. V. (1972). *J. Appl.Phys.*, *43*, 2327.
[8] Khoo, I. C., Kaczmarek, M., Shih, M. Y., Wood, M. V., Diaz, A., Ding, J., Zhang, Y., & Patel, J. (2001). *Mol. Cryst. Liq. Cryst.*, *374*, 315.
[9] Rubinov, A. N. & Efendiev, T. Sh. (1974). *J. Appl. Spectr.*, *21*, 634.
[10] Ilchishin, I. P., Tikhonov, E. A., & Shpak, M. T. (1987). *Sov. J. Quantum Electron.*, *17*, 1567.
[11] Il'ichev, N. N., Malyutin, A. A., Pashinin, P. P., Raspopov, S. F., & Sukhodolsky, A. T. (1982). *Tekh. Phys. Lett.*, *8*, 460.
[12] Bezrodnyi, V. I., Zabello, E. I., & Tikhonov, E. A. (1984). *Sov. J. Quantum Electron.*, *11*, 2438.