

THRESHOLD CONTROL AND LASING CHARACTERISTICS IN DYE DOPED NEMATIC LIQUID CRYSTALS

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Spectral characteristics and lasing efficiency of nematic liquid crystals (NLC) doped with polymethine dyes have been investigated. The lasing threshold control in such materials by variation of the NLC director position relative to orientation of pumping emission linear polarization were obtained. The basis reason of the low lasing efficiency of doped NLC was shown to be the light scattering by the fluctuation of molecular director of the liquid crystal. The rotation of the polarization in such a system following mechanical rotation of the cell with liquid crystal solution is demonstrated experimentally.

I. INTRODUCTION

The investigations of lasing dyes in liquid crystals of various types [1-6] show that doped nematic liquid crystals are prospective materials for lasers whose lasing parameters can be controlled with weak external electric fields. Lasing of dyes in cholesteric liquid crystals (CLC) makes it possible to realize simple and compact lasers with distributed feedback (DFB) [3]. The tunability of lasing in these systems was achieved by exploiting different methods: by varying temperature [3], by applying a mechanical stress [6], by modifying the helical pitch by means of a photo-induced transformation of the chiral dopant [7], by applying an electric field in a waveguide laser [8]. As it is known [9], applying the electric field to a layer of CLC planar texture along the helical axis, along which the lasing emission of the DFB laser is directed, is accompanied by its distortion which heavily decreases the optical transmittance of liquid crystal. Another possible geometry of applying electric field, transverse to the helical axis, is hardly applicable for practical use, first of all due to necessity of applying high voltages (>1 kV).

Up to now, lasing of solutions of dyes in NLC was obtained for a number of polymethine dyes [1], rhodamine 6G [2], coumarine 6 [10], as well as for derivatives of oxazine 17 [11]. The choice of dyes for lasing in NLC is determined mainly by their solubility in such matrix for obtain necessary optical density ($D = 4 \div 6$) in the layers with 200 - 400 μm thickness and rather high measure of ordering of their molecules in NLC. The latter property is determined by the anisotropy of spatial structure of the dye molecule. Among known lasing dyes, the greatest anisotropy have polymethine dyes, for which lasing in oriented liquid crystal was obtained for the first time [1]. Up to now, however, the data on the lasing efficiency in oriented NLC, both polymethine dyes and the dyes of other types [10-11], are absent in the literature. There are a little data on the spectral regions of lasing of doped NLC, and there is only one paper [10] on the change of the lasing characteristics of such lasers when rotating of the NLC director with respect to the direction of pumping polarization. Recently [12-13] it was shown that the doped NLC is able of lasing, including encapsulated in polymer films, have prospects to be used in the systems of displaying of information, as the elements of color laser projection screens, whose optical characteristics are controlled with weak external electric fields. This confirms the urgency of investigations of lasing characteristics of such materials.

In this paper we present the results of investigations of spectral characteristics and lasing efficiency of NLC doped by polymethine dyes, as well as of the control of their threshold power excitation when changing the LC director position with respect to orientation of the exciting radiation linear polarization.

II. EXPERIMENTAL TECHNIQUE

We employed a nematic matrix consisting of ZhK-654 multicomponent mixture with the temperature range of mesophase $-10 \div 65^\circ\text{C}$ and n-methoxybenzylidene - n'-butylaniline (MBBA) with the corresponding temperature range $16 \div 42^\circ\text{C}$. The NLC were mainly activated with indolenone dyes of polymethine class, which typical structural formulae are given in [14]. We used dyes synthesized in the Institute of Organic Chemistry of the National Academy of Sciences («for QE»), without an additional purification. The dye concentration varied from 0.3 to 0.5%, depending on the type and thickness of the NLC layer. The NLC planar texture with optical axis parallel to the substrate plane was formed between glass substrates with a SnO_2 layer. A layer of polyimide lacquer with a thickness not more than $1\mu\text{m}$ was applied in one direction above the SnO_2 layer. Such coating improved the quality of the planar texture [15] and served as transparent electrodes in the case of necessity of applying an electric field to the NLC layer. Thickness of the NLC layer was controlled with fluoroplastic spacers within $200 - 400\mu\text{m}$. Absorption spectra of impurity NLCs were measured with a SF-20 spectrophotometer.

Experiments were performed with the use of a laser system similar to that described in [16]. A NLC laser was pumped by a ruby laser ($\lambda_{\text{ex}} = 694\text{ nm}$) with passive Q-switch oscillating in a single-pulse mode. The energy of pumping radiation and that of lasing of doped NLC was measured with an IMO-2 calorimeter. The pumping radiation was adjusted with neutral optical filters. The lasing spectra were recorded by a spectrograph which included a tube UF-85 and a diffraction grating (1200 ruling/mm, inverse dispersion 0.6 nm/mm in visible range). The cell with doped NLC was placed in the resonator with reflectance of mirrors equal to 60% and 100%. We used almost longitudinal excitation, the minimal angle between the pumping radiation and lasing of the doped NLC was 6° (Fig.1). For maximal use of the dichroism of absorption of lasing dyes, the cell was installed parallel to the resonator mirrors, and we increased the density of power of pumping radiation with the use of a telescopic system.

III. DATA AND DISCUSSION

For anisotropic solution of dichroic dye in NLC, the orientational distribution of their molecules similar to that for the molecules of liquid crystal causes the dependence of their absorption cross-section from the main electronic state on the angle between direction of the incident light radiation polarization and the director (optical axis) of the doped NLC planar texture. Such change in the absorption cross-section of molecules in oriented solution at rotation of the NLC director with respect to the pumping radiation polarization causes changes in the optical density of the solution.

Fig.1 presents the experiment scheme for the threshold control of lasing in doped NLC. The original orientation of the director \mathbf{n} of liquid crystal in the cell C is parallel to the vector \mathbf{E} of the pumping radiation. The orientation of the \mathbf{E} vector of lasing radiation is also vertical. At mechanical rotation of the cell and, correspondingly, of the liquid crystal director \mathbf{n} from its vertical orientation by 84° with respect to the orientation of the pumping vector \mathbf{E} , the lasing radiation vector \mathbf{E} rotates synchronously as well.

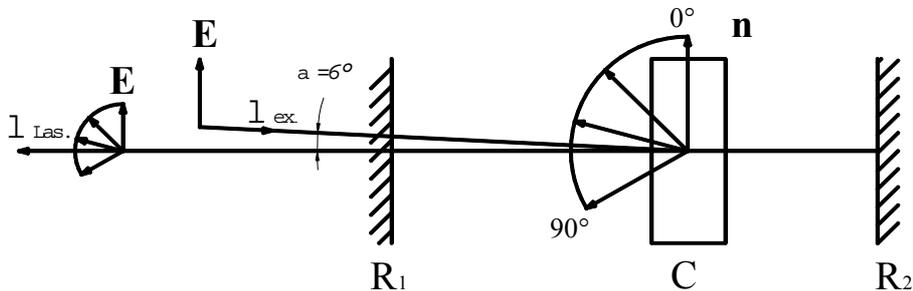


Fig.1: Scheme of experiment on the lasing threshold control in doped NLC. The orientation of E vector of pumping is vertical. The orientation of the NLC director \mathbf{n} varies from vertical to horizontal by 84° angle with respect to polarization of pumping. The vector E of the lasing radiation changes synchronously with the orientation of NLC director. Reflectance of mirrors of resonator; $R_1 = 60\%$, $R_2 = 100\%$.

Fig. 2 shows the dependence of the optical density (D) of the dye PD-3 (Table 1) in NLC ZhK-654 on the mutual orientation of the incident light linear polarization and the liquid crystal director is presented. One can see that, at their parallel orientation, the optical density of the solution is maximal, and it is correspondingly minimal at perpendicular orientation. The value of observed dichroism of optical absorption of the dye characterizes its ordering in the nematic liquid crystal [9]. For PD-3, for which the spatial structure of the molecule is the most anisotropic among the polymethyne dyes which we studied, the ordering parameter in ZhK-654 is equal to 0.56.

One should note that both mechanical rotation of the cell with NLC with respect to the direction of the linear polarization of the incident light and the rotation of the director of the liquid crystal by 90° by means of applying electric field influences the optical absorption in the same way [9]. Installing such a cell with the lasing dyes in resonator at excitation with polarized light must lead to the changes in the optical density of the solution at the frequency of excitation, and it correspondingly changes the threshold density of the pumping power. At constant energy of exciting radiation this leads to changes in the threshold excess number and of the lasing energy.

Figure 3 shows dependence of the pumping power threshold on the angle between NLC director and the direction of exciting radiation polarization for the dye PD-3, whose absorption dichroism is shown in Fig. 2. One can see that the threshold excitation intensity increases from its minimal value, 0.5 MW/cm^2 , by 60 times when turning director \mathbf{n} from 0° to 84° . Optical density of the solution in this case changed from 3.1 to 0.675; higher concentrations were not accessible due to not enough high solubility of the dye. For the others doped NLC, the change in the threshold intensity of excitation was proportional to the linear dichroism of the dye absorption.

The obtained experimental data undoubtedly indicate the absence of the orienting action of the pumping light wave field in the conditions of a single laser pulse. As it is shown in [17], the orientation of NLC during action of a single nanosecond laser pulse ($\tau \approx 15 \text{ ns}$) takes place at intensities of excitation $\approx \text{GW/cm}^2$, and its decrease can be achieved under the action of a series of exciting laser pulses. The orienting action of the laser field for nematic phase is efficient for extraordinary polarization of pumping radiation and rather great angle of incidence at NLC [18]. Such exotic conditions were not realized in our experiments with lasing of doped NLC.

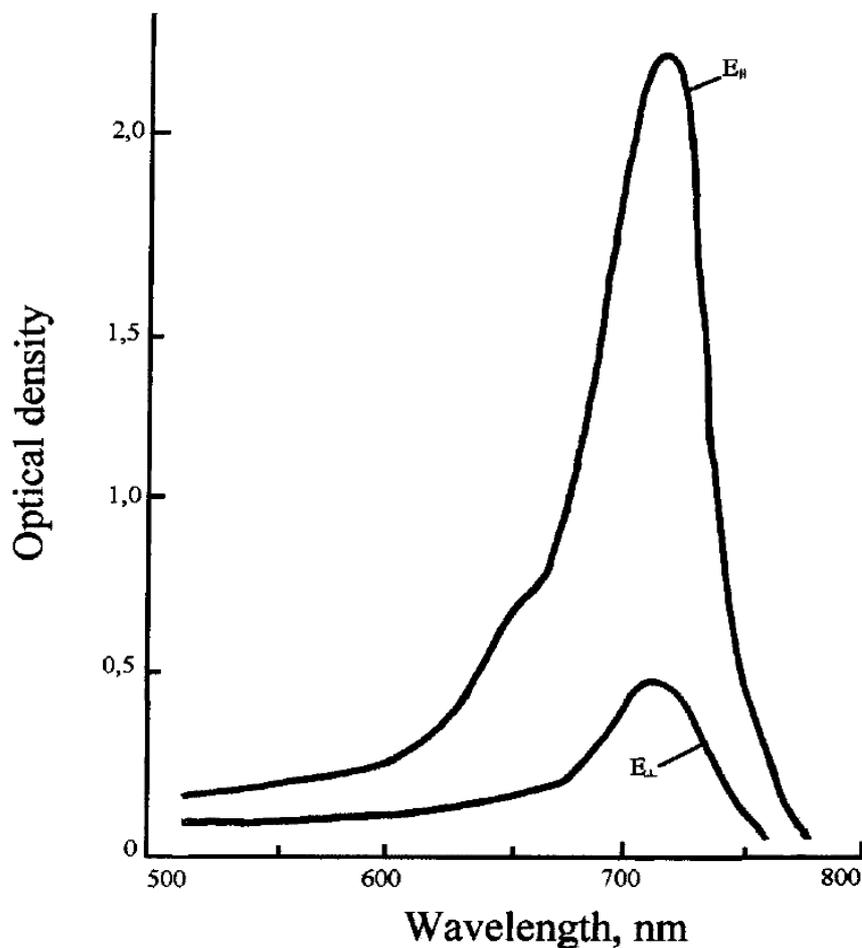


Fig 2: Dichroism of optical absorption for dye PD-3 in ZhK-654. $E_{||}$ and E_{\perp} are parallel and perpendicular orientation of the NLC director with respect to the direction of linear polarization of incident light, correspondingly. The layer thickness is 300 μm .

However, at the pumping radiation intensity of $> 20 \text{ MW/cm}^2$ and optical density of the dye $D=4 \div 5$, one observed long-time (several days) change of the oriented NLC planar texture. In the irradiated zone, there appeared local disoriented planar texture limited with disclination lines. The disorientation disappeared at heating of the sample up to isotropic state and consequent cooling to room temperature ($T=18^\circ\text{C}$). Such disorientation can be caused with the shock wave formed at absorption of intensive pulse of laser radiation. Similar results were also observed in [19] when writing holographic gratings in NLC doped with dyes.

As it is known [1,10], as a result of preferential amplification of the dye fluorescence radiation polarized along the director of NLC, the lasing of dye in such solution is completely linearly polarized, and the direction of polarization at linear polarized excitation (the more so at non polarized) is determined by the NLC director orientation. Synchronous rotation of the polarization plane in the laser on the basis of NLC doped with dichroic dye at rotation of the liquid crystal director with respect to the direction of linear polarization of pumping causes constant level of losses in the region of the lasing frequency. For this reason, notwithstanding the changes in the optical density of the doped NLC on the frequency of excitation at rotation of the cell, which contains it with respect to the vector \mathbf{E} of the exciting radiation, the average frequency of lasing in such a laser does not change.

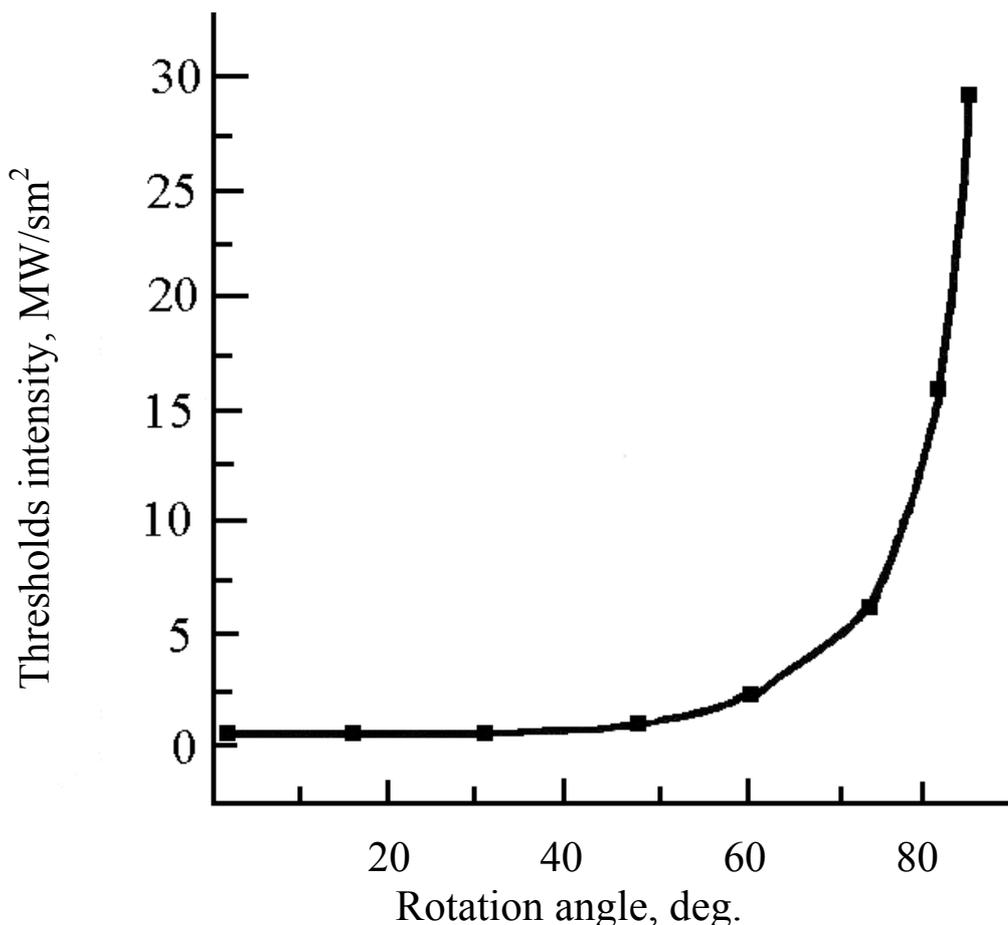


Fig 3: Dependence of threshold intensity of excitation on the angle between of the NLC director position and the direction of linear polarization of exciting radiation. Dye PD-3 in ZhK-654, the layer thickness is 300 μm.

The measurements of the lasing efficiency of NLC doped with polymethine dyes which shows lasing in the 720-830 nm region were performed in the same resonator which was used for the control of the lasing threshold. The cell with doped NLC was installed parallel to the resonator mirrors, the position of the NLC director **n** was set parallel to the **E** vector of the pumping radiation. In Table 1 we present the average values of the lasing efficiency obtained as a result of the series from 3-5 measurements on IMO-2 calorimeters. The data on the lasing efficiency are given without taking into account the losses on Fresnel reflection from the surfaces of cells with doped NLC and incomplete absorption of the pumping radiation.

For most doped NLC given in Table 1, the intensity of excitation equal to 15 MW/cm² exceeded, more than an order of magnitude, the threshold excitation intensities which were equal to 0.5 - 0.7 MW /cm² for the dyes with absorption maxim near 694 nm and 1 - 3 MW /cm² for longer wave dyes. As one can see in Table 1, the lasing efficiency for all dyes is almost one order of magnitude less than in isotropic ethanol solutions (relative lasing efficiency $\eta = E_{\text{las.}} / E_{\text{ex.}} \approx 40\%$). The highest relative lasing efficiency is obtained for NLC doped with PD-3 dye which optical density was close to optimal for the given parameters of resonator. Achieving optimal optical densities of the rest of the dyes was limited by their insufficient solubility in NLC.

An evident reason of the strong decrease of lasing efficiency of dyes in NLC is the losses connected with the Rayleigh scattering caused by thermal fluctuations of director which is 5-6 orders stronger than scattering in isotropic liquids. This is demonstrated by comparison of the lasing

Table 1: Spectral and energy lasing characteristics of a dye doped nematic liquid crystals

Dye	NLC matrices	λ abs., nm	Optical density on frequency excitation	Layer thickness, mm	Lasing band, nm	Relative lasing efficiency η , %
PD-1	ZhK-654	692	2,8	0,3	726-734	2,0
PD-2	MBBA	705	3,5	0,3	728-737	2,8
PD-3	ZhK-654	715	4,2	0,4	747-758	3,4
PD-4	ZhK-654	775	1.6	0,4	810-822	1,2
PD-5	MBBA	700	2,4	0,2	730-738	0,7
PD-5	MBBA (isotropic)	700	5,0	10	736-746	3,8

efficiency of the PD - 5 dye in oriented MBBA and in its isotropic phase at identical conditions of excitation. We observed more than 5 times decrease of the lasing efficiency in the oriented state, while the difference in the thresholds of lasing was little (0.4 - 0.7 MW /cm²). The temperature of isotropic MBBA was 48°C, which excluded any influence of the increase of orientation fluctuations at temperature change of phase on the properties of lasing.

The lasing spectra of dyes in oriented NLC are similar, in their width and structure, to their lasing spectra in isotropic solvents. The structure of the lasing bands is determined by the interference effects on the plane-parallel walls of the cells and in the layer of the doped NLC itself, which is evident from the correlation between the period of this structure and the layer thickness .

CONCLUSIONS

The investigations of the energetic characteristics of the lasers bases on doped NLC shows that the main cause of the lasing efficiency decrease of the activating dyes is scattering caused by thermal fluctuations of the NLC director. Low solubility of the used dyes in NLC hinders one in achieving optimal optical densities even in the layers with 400 μm thickness.

A real way for improving the energetic efficiency of the lasers based on doped NLC could be the increase of dichroic polymethine dyes solubility up to the values which would make possible the use of layers of NLC with the thickness 40-50 μm . This would lead to one order decrease of scattering per single passage through the resonator. Also purpose-oriented synthesis of polymethine dyes with bulky substituent group at nitrogen atoms of chromophore groups of the molecules is necessary which could hinder their coming together in concentrated solution and forming «sandwich» associates.

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