Lasing in the dye doped nematic liquid crystal at the dynamic distributed feedback

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Lasing in the dye doped nematic liquid crystal (NLC) at the dynamic distributed feedback (DDF) induced in the active medium by counter propagating beams is presented. Estimates of the contribution of amplitude and phase gratings to the forward-backward wave interaction in arising periodic structure are made. The narrow frequency tunable emission against the background wide spectrum of the super luminescence is gained, manifesting the oscillation under DDF. Opportunities of control and increase of contrast relation for laser emission are discussed.

I. Introduction

The natural spiral structures of the cholesteric liquid crystal (CLC) allows due to their bulk orientation and dye activation to fabricate the small lasers with the distributed feedback [1-2,4-7]. In ordered planar texture CLC the distributed feedback is caused by circular polarization light Bragg diffraction at the amplitude-phase grating formed by helical structure (a phase grating) and gain of partially ranked dissolved dye (a amplitude grating). For achievement of oscillation with laser pumping to an absorption band of dye the arising gain spectrum should overlap the region of the Bragg diffraction of light with circular polarization at such helical periodic structure.

For the first time the similar laser has been realized in [1]. As a laser medium authors applied derivatives of cholesterol for which the strong dependence of the helix pitch on temperature is characteristic. For thickness of films ≤100 μm sufficient for lasing at their ordering by rubbing the substrates, a relatively weak coupling CLC with the substrates is realized. At the weak coupling change of a helical period with temperature was monotonous as it was required for smooth tuning of lasing wavelength [1].

Since the lasers based on doped CLC compose a unique class of lasers, the active medium of which can be implemented as a surface of any area and curvature without need of the external mirror cavity, they attract great interest in the domain of the development of displays with enhanced brightness and color laser projection screens [3].

Recently examinations of such lasers with application of new, more technological liquid crystal materials have begun [4-7]. Temperature tuning of an oscillation frequency of the CLC lasers is quite inertial, so it needs simultaneously the thermostable operation condition, that appreciably restricts its possible application. Photoinduced change of a period of CLC spiral as an expedient method of an frequency tuning of such lasers, studied in elaborations [8-12] can not be viewed practically important because of difficulties of a step reverse of a spiral after an irradiation. Deformation of the extension of the polymeric CLC as an expedient frequency tuning [6] can only be applied to this type of lasers. Therefore for lasers of such a type there is an actual search and development of more practical methods of oscillation frequency tuning.

The purpose of the present work is studying the impurity nematic liquid crystal laser based on the dynamic periodic structure photoinduced by laser beam pumping in NLC. Such a laser in the short term will allow to apply an electric field for the oscillation frequency tuning of the bulk NLC lasers as it was already made for its waveguide doubles [7].

II. A justification of the approach

The aim of our approach is to create a DDF NLC laser with spatial dynamic structure induced by the interference of the incoming and reflected pumping beams (Fig.1). Usage of the impurity NLC with the positive dielectric anisotropy in such a planar cell when its optical axis is parallel to surface of the substrates and apposition of the electric field between the substrates makes it possible to change a slope angle of an crystal optical axis and to change correspondently the averaged index of refraction of a mesomorphous laser media. It results in the electric field control of the grating spatial period and the lasing wavelength in concordance with expression (1) below.

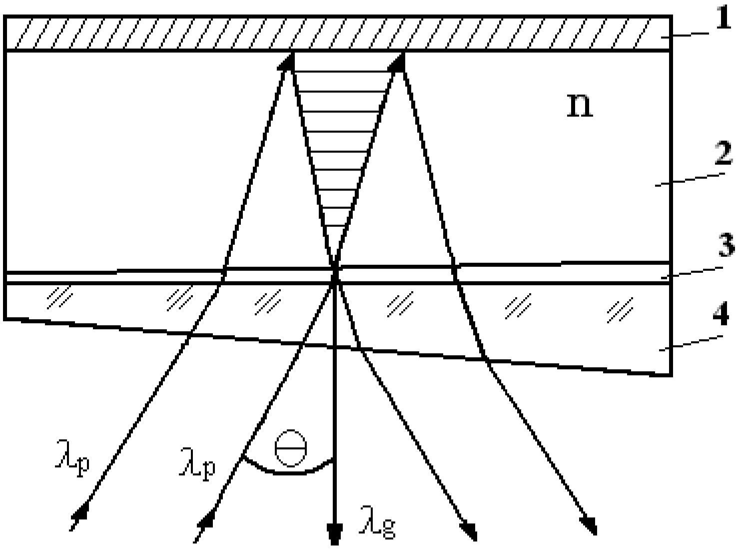


Fig. 1: A configuration of the laser cell. 1 – light reflecting substrate with an ordering film (polyimide), 2 –NLC layer, 3 – the transparent SnO_2 electrodes, 4 – glass substrate

For given scheme of the laser pumping the photoinduced phase planes are parallel to both substrate surfaces and the oscillation wavelength of the DDF NLC laser is determined by the incidence angle of a pumping beam θ and the index of refraction (n) of the NLC active medium [13-14], namely

$$\lambda_{g.} = \lambda_p \, n / (n^2 - \sin(2\theta))^{1/2} \qquad (1)$$

here - $\lambda_{g.}$ and λ_p is lasing and pumping wavelength.

Above presented configuration of DDF NLC lasing allows to realize the unidirectional output of laser emission that is quite useful for application.

Let's estimate some parameters of an amplitude-phase grating which can take place in our experiment conditions. We shall estimate the comparative contributions of an amplitude grating of amplification due to dye excitation and a phase grating due to a spatially modulated absorption and heating in the interaction power of counter running waves in periodic structure.

According to the theory of distributed feedback lasing in an isotropic material [15], the interaction power of waves in such a structure is defined by a product of an interaction constant χ and length of the structure L. In the case of amplitude-phase structure aroused at an interference of pumping beams , the interaction constant equals:

$$\chi = \pi\ \Delta n/ \lambda + i\ \Delta\alpha/2 \qquad (2)$$

Here Δn is the depth of modulation of the index of refraction of NLC due to heating and other nonlinear interaction, $\Delta\alpha$ is the depth of modulation of the gain due to population inversion of dye molecules, λ is the wavelength corresponding to the Bragg conditions. To compare the mutual contribution of each process in the conditions of a real experiment we shall rewrite down a square of the module of the constant:

$$|\chi|^2 = (\pi\Delta n/ \lambda)^2 + \Delta\alpha^2 /4 \qquad (3)$$

For estimates of both terms of this equation we use parameters of DFL with a dynamical amplitude-phase grating on the basis of a rhodamine 6G solution in ethanol. In typical requirements of experiment for the DFL $\Delta\alpha \approx$ (4-5) cm^{-1} [16]. For the estimation of the magnitude $(\pi\Delta n/\lambda)^2$ it is necessary to know the value of amplitude of modulation of the index of refraction of NLC.

Change of the refraction index under the power light illumination occurs at participation of various mechanisms: Kerr effect, electrostriction and the thermal heating of media:

$$n = n_0 + n_2 E^2 \qquad (4)$$

The second term in (4) is the change of the index of refraction due to the light intensity only. For the typical for lasing of dye ethanol solution pumping power the nonlinear makeweight n_2E^2 due to electrostriction and Kerr effect equals approximately 10^{-9} and 10^{-10} correspondently [17].

Under direct light absorption as it works at dye laser pumping the variation of the index of refraction can be estimated by means of the thermodynamical consideration and the relation:

$$\Delta n_t = (dn/dT)_p\, \Delta T \qquad (5)$$

where ΔT – change of temperature, $(dn/dT)_p$ – temperature dependence of the index of refraction at abiding pressure. Rise of the temperature ΔT without dissipation (for nanosecond time interval) is equal:

$$\Delta T = Q/ Vc_v\rho \qquad (6)$$

Here Q – the quantity of heat exuded in a pumped region of a dye solution, c_v – specific heat capacity at constant volume, V – volume of an active pumped media, ρ -specific density of a solution. The quantity of heat which exuded in the pumped area of a dye solution during a laser pulse is approximately defined by [18]:

$$Q = E_p\ (1- \eta)\ \lambda_{p/}\lambda_g \qquad (7)$$

Where E_p-energy of a pumping pulse, λ_p, λ_g – wavelengths of pumping and oscillation emission, η - lasing efficiency . At the used values of pumping and thermodynamical solution parameters (E_p =3 mJ, λ_p =530 nm, λ_g = 560nm, $_{Cv}$ =2,43 J/gT, $V = 5\times10^{-5}$ cm^3, ρ = 0,79 g/cm^{-3} , $(dn/dT)_p = - 4 \cdot 10^{-4}$) we gain value $\Delta n_t \approx 10^{-4}$ and $(\pi\Delta n/ \lambda)^2 \approx 25$ cm^2.

At comparison of contributions into a feedback power of a amplitude (gain) part and a phase part of grating the prevailing weight of a phase grating and including its thermal builder is evident. Threshold pumping power for DFL laser on the basis of cholesterol derivatives for which the theory of DFL [15] is valid, to average 50-100 kW/cm^2 at Δn $5\cdot10^{-2}$ (birefringence) and layer thickness about 50 μm [19]. For supposed DDF NLC laser at $\Delta n\approx10^{-4}$, lasing conditions are appreciably more complex in comparison with the similar laser on steroid CLC [19], therefore for increase of interaction power the thickness of an active layer needs to be taken greatest possible.

III. Technique of experiment

For the fulfillment of necessary requirements for the laser oscillation we compared functioning DDF laser in above proposed configuration in the thin films of isotropic and anisotropies media. As an isotropic active medium the rhodamine 6G ethanol solution was taken. As an anisotropy media – solutions of the different class dyes in NLC ZhK-654, representing a commercial composition of mesomorphous material with the positive dielectric anisotropy were used. We measured threshold and spectral characteristics of DDF laser on these media depending on thickness of layer and their optical density at excitation by second harmonic of a Nd^{3+} laser with passive Q-switch oscillating. Thickness of the active layers was defined by spacers of a cell and was in boundaries of 250 μm ÷ 1 mm for isotropic, and 250 μm – for ordered dye doped NLC. The scheme of the experiment is presented on fig. 2.

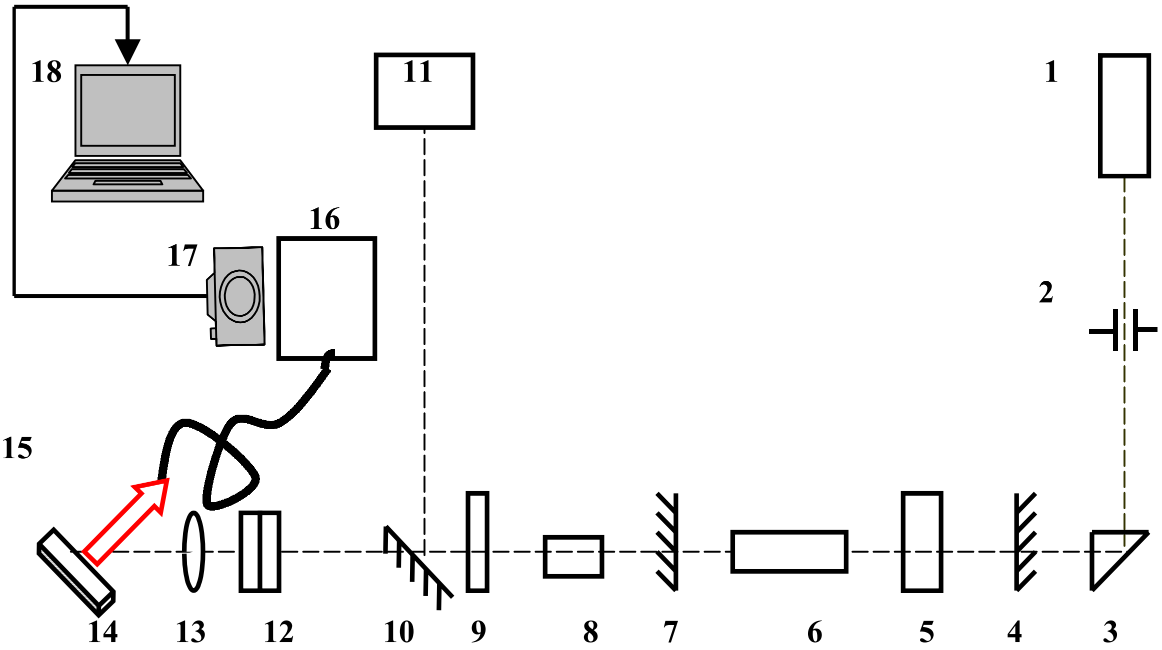


Fig. 2 : Experimental setup. 1 – He-Ne laser, 2- stop's screen, 3- TIR-prism, 4,7 – mirrors, 5 – passive Q-switch modulator, 6 – Nd^{3+} laser, 8 - KDP crystal, 9 - selective filter, 10 – dichroic mirror, 11 – calorimeter, 12 – neutral filters, 13- focusing lens, F=21sm, 14- cell with active medium, 15- optical fiber, 16–spectrograph, 17 - web-camera, 18 - personal computer.

The second harmonic of the laser radiation 532nm, $\tau \approx 20$ns was focalized by a lens on a cell with a dye solutions. To decrease a stray oscillation due to an external cavity one of the substrates is made as a wedge with an angle of 3 degrees. In experiments with the impurity NLC the interior surfaces of both substrates shown in fig.1. have been coated by a polyimide films to order a mesomorphous media by their subsequent rubbing. Spectrums of lasing were registered by a web-camera in a focal plane of a spectrograph (inverse dispersion 0,6 nm/mm), with their further computer processing.

IV. Results and discussion

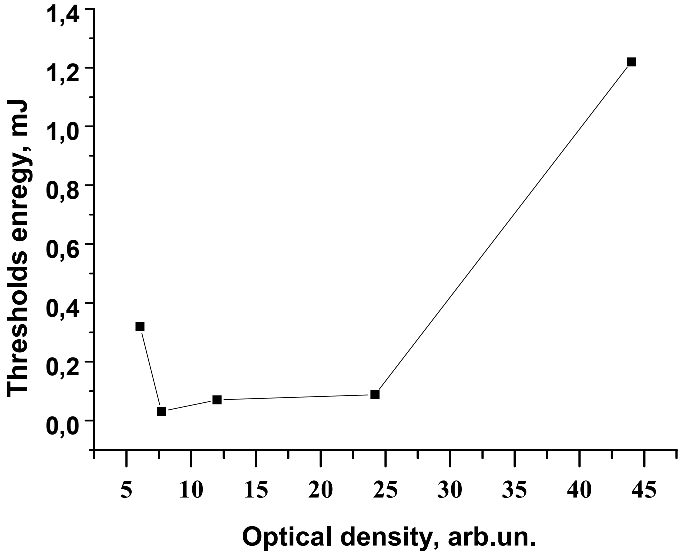
Performances of DDF laser with counter propagating beam pumping with isotropic media were earlier studied in [13-14,20] at thickness of the active media more than 1 mm. In these operations wedge-like layer of the active medium were used. For DDF NLC laser it was necessary to use a parallel layer of the active medium as in wedge-like layer there is a variable declination of an optical axis at superimposition of an electric field and different frequencies of oscillation in an excitation band are accordingly possible

In fig.3 the dependences of the threshold pumping energy of DDF laser versus an optical density of a R6G solution are presented at various layer thickness. Apparently thresholds of oscillation are high enough in comparison with similar thresholds of oscillation of DDF R6G laser, which are realized in volumetric layer (thickness more 1mm) [16]. From the given dependences it is clear that for the active layer thickness of 250 μm the optimum value of the optical densities makes from 3 up to 6. Such layer thickness should be used for reception of a mode of oscillation of the impurity NLC.

For layer thickness of 400 μm we studied spectrums of oscillation of DDF laser depending on an incidence angle of pumping beam. It has been found that the spectral width makes 1,8 nm at 5 - multiple pumping-over the threshold while the spectral width the same dye solution in a usual 5mm cell with cavity formed by walls makes 9nm at similar pumping-over above a threshold. At change of an incidence pumping beam angle in limits 27 ÷33 degrees on a cell with mirror reflecting substrate the frequency tuning in a boundary 560 - 561,5 nm has been observed.

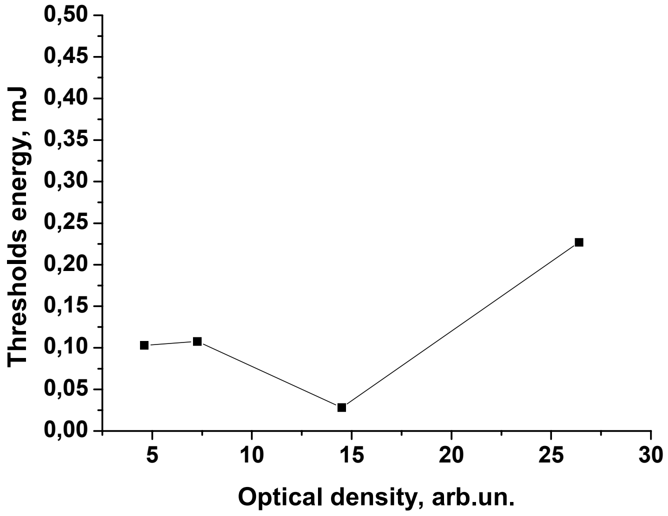
These data have been used to fabricate the following DDF NLC lasers. For the first time as doping laser dyes for NLC ZhK-654 the charge neutral and spatially isotropic dyes and the ionic polymethyne spatially anisotropic dyes were used. The scheme of experiment was the same as at study of the isotropic solutions. Pumping beam polarization was vertical, an optical density at 532nm was about 3. Unfortunately the lasing threshold for this dyes in NLC ZhK-654 in all gamut of pumping power up to 40 MW/cm^2 has not been reached. The most probable reason of failure was concentration quenching of fluorescence quantum yield down to a level of 1-3 %. The same failure was for the case of polymethine dye doping when the quantum yield was higher (10%).

The suitable dye for laser oscillation in NLC ZhK-654 at the impulse excitation by 532nm has been found among the pyrromethene laser dyes. These dyes are characterized by a high solubility in NLC and concentration quenching deficiency at all used concentrations. The NLC solution of pyrromethene dye No 567 with a maximum of absorption and fluorescence at 524nm and 548nm has quantum yield 98%.



a – 1mm

Thresholds enregy, mJ
1,4
1,2
1,0
0,8
0,6
0,4
0,2
0,0
5
10
15
20
25
30
35
40
45
Optical density, arb.un.



b-650μm

Thresholds energy, mJ
0,50
0,45
0,40
0,35
0,30
0,25
0,20
0,15
0,10
0,05
0,00
5
10
15
20
25
30
Optical density, arb.un.

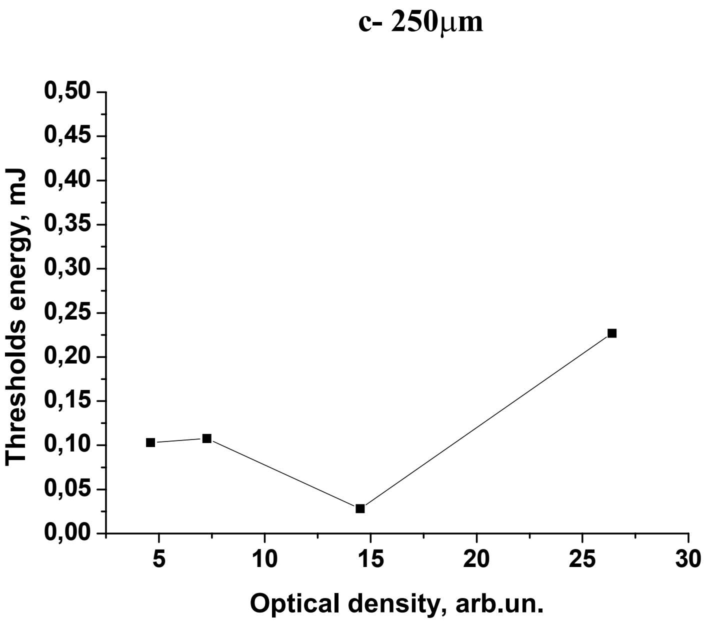


Fig.3: The dependence of lasing thresholds energy of DFB-laser based on isotropic solution as function of optical density on pumping wavelength for 3 thickness of active media.

In fig.4 absorption spectra of this dye in NLC ZhK-654 are presented. As it seen the absorption differs for the linear polarization of incident light parallel to director of NLC (a curve 1) and for perpendicular orientation – (a curve 2). The relatively low dichroism of absorption hints about the small anisotropy of its spatial structure. Calculated from fig.4. the order parameter of pyrromethene dye No 567 in NLC ZhK-654 makes ≈ 0,13.

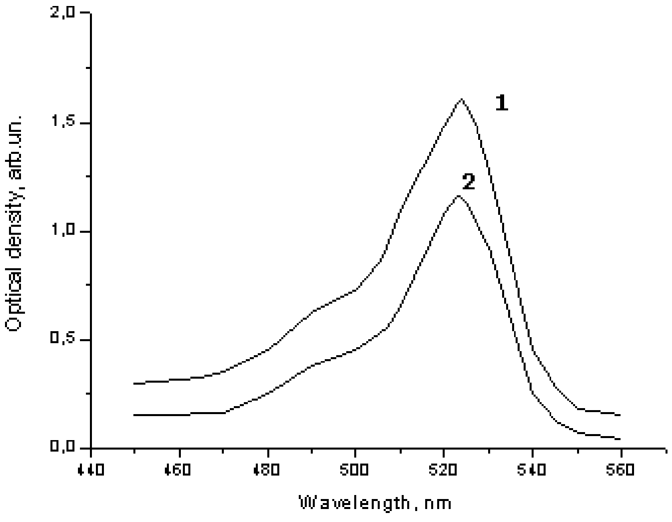


Fig.4: Absorption spectra of pyrromethene dye No 567 in NLC ZhK-654: 1- light polarization is parallel to director , 2 – light polarization is perpendicular to director.

With this dye solution in NLC ZhK-654 under the scheme of excitation by the colliding beams, presented in fig.1. laser oscillation has been realized**.** The layer of the impurity NLC with the thickness 250 μm and an optical density at a wavelength 532nm equaled 3 was used. The oscillation threshold of laser is receipt the same level, as for the R6G ethanol solution. In fig.5 a lasing spectrum of the dye doped NLC ZhK-654 is presented. The laser emission was registered by means of a web-camera from the matte plate placed in the focal plane of a spectrograph at the pumping beam incidence angle 35°. The medial wavelength of lasing makes 554 nm, at total spectrum width about 4 nm.

Apparently from the presented picture against the background of a diffuse spectrum of 4nm the narrow intensive line with a wavelength ≈ 553,3 nm is shown. Study of the emission spectrum at variations of a pumping beam incidence angle have shown, that a narrow line arises only in the certain gamut of excitation angle (28-37°). Frequency dependence of the narrow oscillation line from the pumping beam angle testifies to laser oscillation in requirements of DDF on impurity NLC simultaneously with the stray super luminescence caused by reflection from a mirror. Suppression of the broad super luminescence emission and increase of contrast relation of the lasing line is the content of the further development in the given problem. The low threshold of super luminescence in the given configuration (fig.1.) is an uneasy problem, particularly in connection with introduction of the transparent electrodes made of SnO_2 on an axis of oscillation. Though reflection from the transparent substrate at presence of stratum NLC makes 5-6 %, however super luminescence in such configuration becomes the important competing process because of a high oscillation threshold of main cavity – DDF structure for a such scheme. Increase of a oscillation threshold of DDF laser in above used configuration caused by that the second pumping beam responsible for interference is attenuated owing to passage through absorbing active layer. The interference of two beams strongly differing in intensity and a degree of a spatial coherence leads to depression of contrast, i.e. diminution of a depth of modulation of the peak-phase grating responsible for efficiency of DDF that deteriorates lasing conditions.

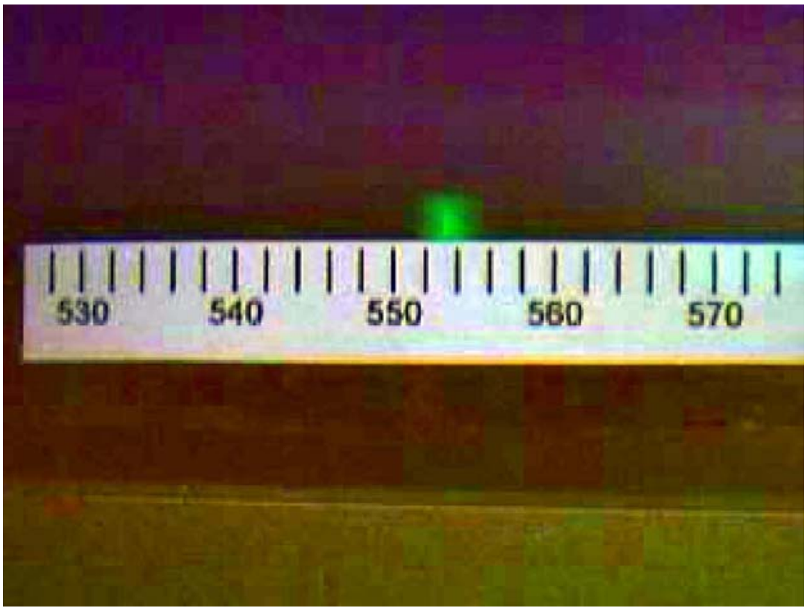


Fig. 5: A lasing spectrum of pyrromethene dye No 567 in NLC ZhK-654.

The increase of a modulation depth (or grating strength) is possible to reach by means of reducing an optical density of the doping dye, however that will results the higher threshold DDF lasing because of decrease in amplification.The best results is expected at the usage of the single-mode pumping beam with a 100% spatial coherence over the whole cross section. More cardinal solution concerning increase of contrast interference pattern in pumping and the convenient control of laser parameters will be reached with application DDF lasing by an interference in passing beams and with application the traversal electric field to a direction of oscillation emission.

V. References

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