

LASER APPLICATIONS AND OTHER TOPICS IN QUANTUM ELECTRONICS

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Influence of the layer thickness and concentration of dye molecules on the emission amplification in cholesteric liquid crystals

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***Abstract*. The propagation of light through a planar layer of a cholesteric liquid crystal doped with dye molecules is considered. The features of the emission spectra of the crystal are studied both in the absence and presence of dielectric boundaries. The increase in the emission intensity is investigated for different layer thicknesses and different concentrations of dye molecules. It is shown that an anomalously strong increase in the emission intensity with the diffraction intrinsic polarisation takes place in the case of a comparatively small crystal thickness and a relatively low concentration of dye molecules. The obtained results can be used for the development of miniature lasers with the circular polarisation of the fundamental radiation mode.**

Keywords: *cholesteric liquid crystals, laser action, dye lasers, diffraction reflection and transmission.*

1. Introduction

It is known that liquid crystals (LCs) containing chiral molecules have the self-organising helical structure. The local sites of such a structure represent an anisotropic medium with the optical axis characterised by the unit vector called a director. The director continuously rotates along the spiral axis by forming a twisted birefringent medium. Upon the propagation of light along the axis of the medium, the two eigenmodes excited in it have opposite quasi-circular polarisations. For the eigenmode with the polarisation direction coinciding with the rotation direction of the helix in the medium, the forbidden photonic band exists. Light with such polarisation incident on a layer of the medium experiences diffraction reflection in the wavelength region from $\lambda_1 = \sigma n_1$ to $\lambda_2 = \sigma n_2$ (n_1 and n_2 are the local ordinary and extraordinary refractive indices and σ is the helix pitch). For another eigenmode, the photonic forbidden band is absent and incident light with the opposite quasi-circular polarisation does not experience diffraction reflection.

Theoretical and experimental studies of the optical properties of LCs continue to attract the interest of researchers because the results of these studies can find technical applications in devices of a new generation. Recently chiral LCs (cholesteric LCs, chiral smectics, etc.) and artificial chiral-constructed crystals [1 – 3] find wide applications in highly efficient liquid-crystal displays [4, 5], modulators [1, 2, 6, 7], polarisation filters and mirrors [1, 2, 8 – 10], for imaging in polarised light, as sources of circularly polarised radiation [1, 2, 8, 9], etc. The emission spectrum of amplifying media [in particular, cholesteric LCs doped with molecules with the fluorescence band located in the diffraction reflection region (DRR) or including it] considerably depends on their periodic structure. In such media, low-threshold narrowband lasing without mirrors can occur at the DRR edges upon optical pumping [11 – 16]. The observation of low-threshold lasing in cholesteric LCs (CLCs) at the DRR edges rather than at the DRR centre was explained only recently by the fact that the photonic density of states tends to zero in the DRR and has sharp peaks at the DRR boundaries [17].

The total electric field in the DRR in a CLC irradiated by light with polarisation coinciding with the polarisation of the diffracted eigenmode has linear polarisation [18, 19] directed along the local director on the long-wavelength boundary of the DRR or perpendicular to it (on the short-wavelength boundary of the DRR). Therefore, as shown in [20], if a high degree of ordering of the transition dipole moments of impurity molecules along (or perpendicular to) the director is provided, the emission intensity at the short-wavelength (long-wavelength) boundary increases, providing the possibility of low-threshold lasing. Because the pitch of the CLC helix can be easily externally controlled, it is possible to tune the laser wavelength, which is very important for different applications. The influence of the electric field, temperature and optical irradiation on the lasing properties of CLCs doped with laser dyes was studied in papers [12], [13, 15], and [14], respectively. The theory explaining the suppression and amplification of circularly polarised emission in CLCs was developed in papers [21, 22]. The lasing properties of CLCs were studied theoretically in papers [23, 24], where the lasing conditions were obtained and the lasing threshold was calculated.

In this paper, we studied the influence of the layer thickness and concentration of fluorescing impurity molecules on the emission amplification in CLCs.

2. Calculation method

Consider the reflection and transmission of light by a planar CLC layer. We assume that CLC molecules are

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perfectly ordered in a helix structure, i.e. the order parameter is $S = 1$. Let us expand the complex amplitudes of the electric fields of the incident, reflected, and transmitted waves in the basis circular polarisations:

$$\boldsymbol{E}_{\rm i,r,t} = \boldsymbol{E}^{+}_{\rm i,r,t}\boldsymbol{n}_{+} + \boldsymbol{E}^{-}_{\rm i,r,t}\boldsymbol{n}_{-} = \begin{pmatrix} \boldsymbol{E}^{+}_{\rm i,r,t} \\ \boldsymbol{E}^{-}_{\rm i,r,t} \end{pmatrix}, \qquad (1)$$

where the indices i, r, and t correspond to the incident, reflected, and transmitted waves; and $\boldsymbol{n}_{+}$ and $\boldsymbol{n}_{-}$ are the unit vectors of the right- and left-hand circular polarisations.

By using the solution of Maxwell's equations for a CLC [2], we will write the solution of the boundary problem for the reflection and transmission of light through a finite CLC layer, representing a system of eight linear equations (continuity conditions for the tangential components of fields at the layer boundaries), in the form

$$\boldsymbol{E}_{\rm r} = \hat{R}\boldsymbol{E}_{\rm i}, \quad \boldsymbol{E}_{\rm t} = \hat{T}\boldsymbol{E}_{\rm i}, \qquad (2)$$

where $\hat{R}$ and $\hat{T}$ are the 2×2 reflection and transmission matrices of the given system. According to [25], the elements of matrices $\hat{R}$ and $\hat{T}$ have the form

$$R_{11} = R_{22} = H, \quad R_{12} = B + {\rm i}F, \quad R_{21} = B - {\rm i}F,$$

$$T_{11} = (S - {\rm i}N)\exp({\rm i}2ad), \quad T_{12} = V\exp({\rm i}2ad), \qquad (3)$$

$$T_{21} = V\exp(-{\rm i}2ad), \quad T_{22} = (S + {\rm i}N)\exp(-{\rm i}2ad),$$

where

$$H = \{\chi^2 r_2 r_1(c_1c_2 - 1) + 2u^2[r_2r_1(2\chi^2m_1 - \gamma^2) - \alpha^2\delta^2\gamma^2]s_1s_2 - {\rm i}u\sqrt{\alpha}\,\gamma(p_1s_1c_2 + p_2s_2c_1)\}\Delta^{-1};$$

$$F = \delta\chi\sqrt{\alpha}\{-2u\gamma\sqrt{\alpha}(s_1c_2 - s_2c_1) - {\rm i}[r_2(c_1c_2 - 1) + 4u^2(m_1r_2 + \alpha\gamma^2)]s_1s_2\}\Delta^{-1};$$

$$B = u\gamma\delta\sqrt{\alpha}[4u\sqrt{\alpha}\,\gamma s_1s_2 + {\rm i}(g_1s_2c_1 - g_2s_1c_2)]\Delta^{-1};$$

$$S = \gamma\sqrt{\alpha}[\gamma\sqrt{\alpha}\,(c_1 + c_2) - {\rm i}u(b_1s_1 + b_2s_2)]\Delta^{-1};$$

$$V = \gamma\sqrt{\alpha}\,\delta[\sqrt{\alpha}\,(c_2 - c_1) - {\rm i}u(q_2s_2 - q_1s_1)]\Delta^{-1};$$

$$N = \gamma\sqrt{\alpha}\,\chi[{\rm i}r_1(c_2 - c_1) - 2u\sqrt{\alpha}\,(l_1s_1 + l_2s_2)]\Delta^{-1};$$

$$\Delta = -\chi^2r_2^2 + (\chi^2r_2^2 + 2\alpha\gamma^2)c_1c_2 + 2u^2[\alpha^2\delta^2\gamma^2 - r_1^2(2\chi^2m_2 + \delta^2) + 4\alpha\chi^2(\delta^2 - 2m_2)]s_1s_2 - {\rm i}2u\gamma\sqrt{\alpha}\,(b_1s_1c_2 + b_2s_2c_1);$$

$$b_{1,2} = r_1w_{1,2} \pm \alpha\delta^2; \quad p_{1,2} = r_2w_{1,2} \mp \alpha\delta^2; \quad q_{1,2} = r_1 \pm \alpha\gamma;$$

$$g_{1,2} = r_2 \pm \alpha\gamma; \quad w_{1,2} = \gamma \pm 2\chi^2; \quad l_{1,2} = \gamma \pm 2; \quad r_{1,2} = 1 \pm \alpha;$$

$$m_{1,2} = 1 \pm \chi^2; \quad s_{1,2} = \frac{\sin(k_{1,2}d)}{k_{1,2}d}; \quad c_{1,2} = \cos(k_{1,2}d);$$

$$k_{1,2} = \frac{2ub^{\pm}}{d}; \quad u = \frac{\pi d\sqrt{\varepsilon_{\rm m}}}{\lambda}; \quad b^{\pm} = (1 + \chi^2 - \delta \pm \gamma)^{1/2};$$

$$\gamma = (4\chi^2 + \delta^2)^{1/2}; \quad \varepsilon_{\rm m} = \frac{\varepsilon_1 + \varepsilon_2}{2}; \quad \delta = \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + \varepsilon_2};$$

ε_1 and ε_2 are the principal values of the local permittivity tensor; $\chi = \lambda/(\sigma\sqrt{\varepsilon_{\rm m}})$; $a = 2\pi/\sigma$; d is the layer thickness; $\alpha = (\varepsilon_{\rm m}/\varepsilon)^{1/2}$; and ε is the permittivity of the medium bordering on both sides with the CLC layer.

By using (1)–(3), we can calculate, for example, the reflection ($R = |E_{\rm r}|^2/|E_{\rm i}|^2$) and transmission ($T = |E_{\rm t}|^2/|E_{\rm i}|^2$) coefficients, the linear and circular dichroism, the optical rotation angle $\psi = (-\arg\varkappa)/2$, and polarisation ellipticity $e = (|\varkappa| - 1)/(|\varkappa| + 1)$, where $\varkappa = E_{\rm t}^{+}/E_{\rm t}^{-}$.

3. Results of calculations and discussion

Consider a CLC layer doped with dye molecules which is located between two semi-infinite media. In the presence of a pump wave, this layer is an amplifying medium, i.e. we have a planar resonator with an active element. The presence of dye molecules in the CLC layer leads to a change in the local refractive indices of the medium. In this case, the imaginary parts $n''_{1,2}$ of the effective local refractive indices $n_{1,2} = n'_{1,2} + {\rm i}n''_{1,2}$ are negative. In the presence of absorption (imaginary parts $n''_{1,2}$ of the local refractive indices of the CLC are positive), the quantity $Q = 1 - (R + T)$ characterises the light energy absorbed in the CLC layer, while the emission of the system in the amplifying medium will be characterised by the quantity $|Q|$ (it is assumed that the dimensionless intensity of the incident light is $I_0 = 1$).

Similarly to the order parameter S characterising the orientation of molecules in nematic LCs or CLCs, we will characterise the degree of ordering of the transition dipole moments of dye molecules by the order parameter $S_{\rm d}$ defined by the expression

$$S_{\rm d} = \frac{3}{2}\langle\cos\vartheta\rangle - \frac{1}{2}, \qquad (4)$$

where ϑ is the angle between the local director and the transition dipole moment of dye molecules. The maximum possible order parameter $S_{\rm d} = 1$ corresponds to the orientation of transition dipole moments strictly parallel to the local director. The value $S_{\rm d} = 0$ corresponds to the isotropic orientation distribution of dipole moments, while the minimal value $S_{\rm d} = -0.5$ corresponds to the isotropic distribution of transition dipole moments in a plane perpendicular to the director. In the linear optics approximation, relations (3) describe both amplification and lasing.

We will study the properties of emission amplification for the waves with intrinsic polarisations. The intrinsic polarisations are the two polarisations that do not change during the propagation of light in a medium [26]. It follows from this definition that intrinsic polarisations should be related to the polarisations of eigenmodes excited in the medium. We calculated intrinsic polarisations taking into account the influence of dielectric boundaries. We will call the intrinsic polarisation of radiation diffracted in the medium the diffracting intrinsic polarisation and the intrinsic polarisation of radiation that was not diffracted in the medium the non-diffracting intrinsic polarisation.

Figure 1 presents the dependence of $\ln|Q|$ on the wavelength for different S_d. The light incident on the system has the diffracting intrinsic polarisation [in this case, this is quasi-right-hand circular polarisation, curve (*1*)] and the opposite non-diffracting intrinsic polarisation [quasi-left-hand circular polarisation, curve (*2*)]. The CLC helix is right-hand. We consider the case of normally incident light. Dependences in Figs 1a–c correspond to the case of the minimal influence of dielectric boundaries, i.e. when $\varepsilon_m = \varepsilon$. Figures 1d–f present similar dependences for the case $\varepsilon = 1$, i.e. when the CLC layer is located in air and the influence of dielectric boundaries is considerable. Figure 1 demonstrates the resonance decrease in $|Q|$ in the DRR for the diffracting eigenmode caused by the diffraction suppression of emission, which is similar to the diffraction suppression of absorption. For $S_d = 1$ ($S_d = -0.5$), emission is almost completely suppressed at the short-wavelength (long-wavelength) DRR boundary. Outside the DRR, the resonance increase in the diffracting mode emission is observed, for which the feedback is provided by the periodic stricture of the medium. For $\alpha = 1$, emission with the non-diffracting intrinsic polarisation is virtually absent due to the absence of feedback, while for $\alpha \neq 1$ the feedback (weak) for this mode is provided by dielectric boundaries, and the emission intensity of this mode also somewhat increases.

Note also that a comparison of curves (*1*) and (*2*) (Fig. 1) with similar curves in the presence of absorption shows that absorption leads to the effective decrease in the layer thickness, while amplification causes the effective increase in the layer thickness.

Consider now the influence of the layer thickness on emission amplification. Figure 2 presents the dependences of $\ln|Q|$ on the reduced thickness d/σ of the CLC layer for different wavelengths of the incident light. The first, second, and third rows correspond to $S_d = 1$, 0, and -0.5, respectively. The first three rows correspond to the case $\alpha = 1$, and the next three rows – to the presence of dielectric boundaries ($\alpha = \sqrt{\varepsilon_m}$). The first column corresponds to the wavelength of the incident light 0.5 μm, which lies far outside the DRR, which covers the wavelength range from 0.6148 to 0.6356 μm. The second column corresponds to the wavelength 0.613 μm lying outside the DRR near its short-wavelength boundary. The third column corresponds to the wavelength 0.615 μm lying within the DRR near its short-wavelength boundary, and the fourth column corresponds to the wavelength 0.625 μm located at the DRR centre.

One can see from Fig. 2 that the dependences of the emission intensity on the layer thickness are different for different wavelengths. We observed the following properties. As expected, in the case $\alpha = 1$ for the incident wave with the non-diffracting intrinsic polarisation [curves (*2*)], the value of $\ln|Q|$ monotonically increases with increasing layer thickness. Within the DRR for the incident wave with the diffracting intrinsic polarisation [curves (*1*)], emission is suppressed similarly to absorption suppression (as d/σ increases, the value of $\ln|Q|$ first rapidly increases and then 'saturates'). An interesting picture is observed near the short-wavelength boundary of the DRR for $S_d = 1$: the value of $\ln|Q|$ first rapidly increases achieving a maximum

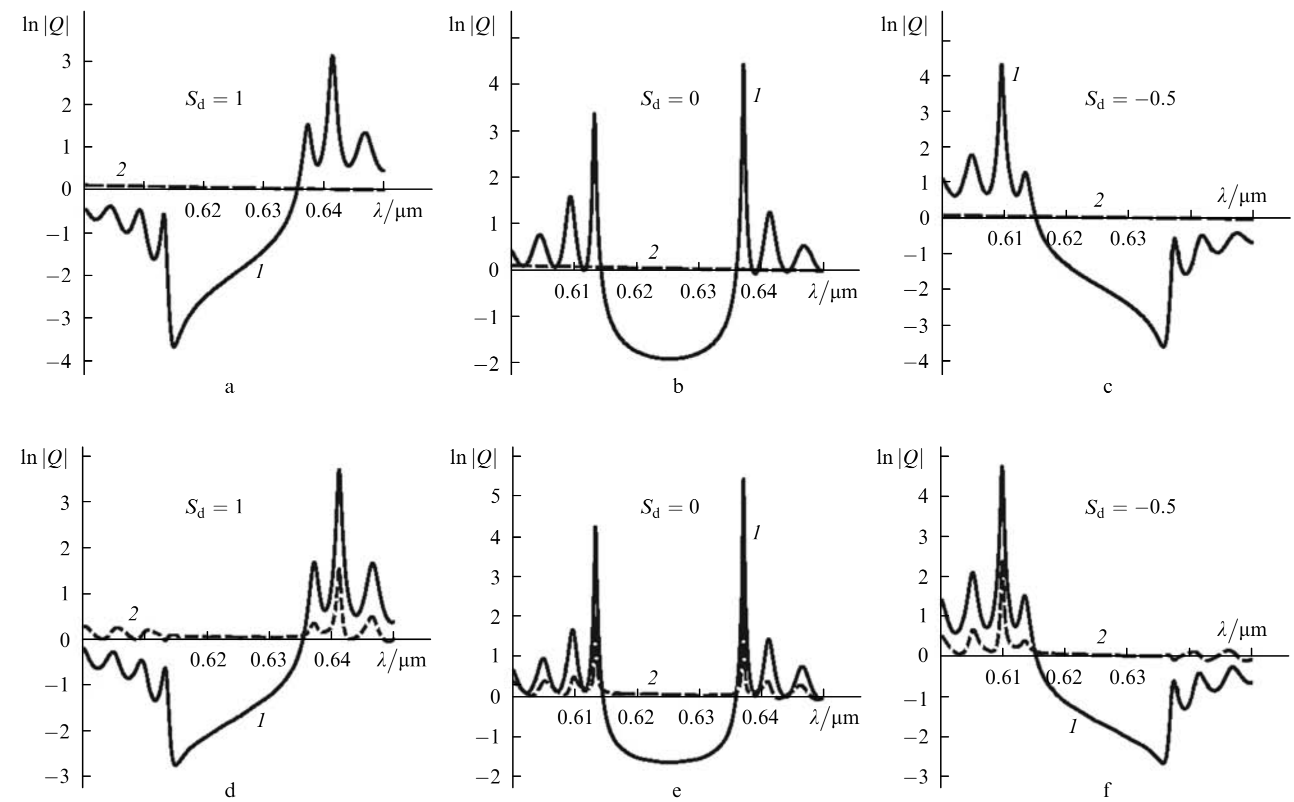

Figure 1. Wavelength dependences of the emission intensity $\ln|Q|$ for different S_d in the absence (a–c) and presence (d–f) of dielectric boundaries. The light incident on a CLC layer has the diffracting (*1*) and non-diffracting (*2*) intrinsic polarisation. The parameters the CLC layer doped with dye molecules are: $\varepsilon_1' = 2.29$, $\varepsilon_2' = 2.143$, $\varepsilon_m'' = -0.005$, $\sigma = 0.42$ μm, and $d = 50\sigma$.

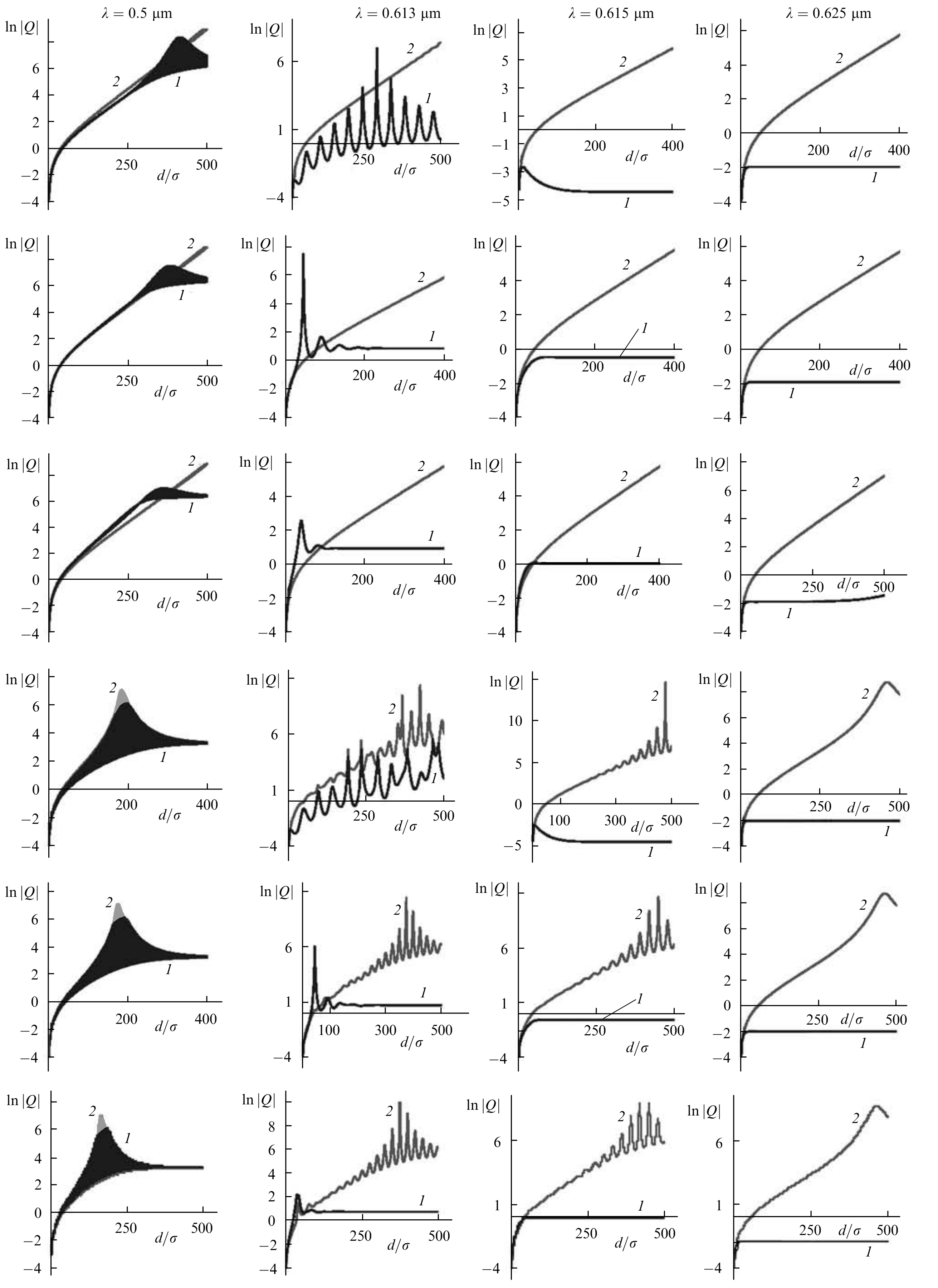

Figure 2. Dependences of the emission intensity $\ln|Q|$ on the reduced CLC layer thickness d/σ for different wavelengths of the incident light in the absence (first three rows) and presence (next three rows) dielectric boundaries. Parameters of the layer and curve numeration are as in Fig. 1.

and then begins to decrease. As the thickness of the CLC layer doped with dye molecules is increased, the emission intensity decreases (of course, in the presence of the pump wave). This effect is analogous to a decrease in absorption in absorbing media with increasing layer thickness [25]. For $S_{\rm d} = 0.5$, a similar effect takes place near the long-wavelength boundary of the DRR. Outside the DRR near its boundaries, an anomalously strong increase in the emission intensity is observed. For $S_{\rm d} = 1$, the value of $\ln|Q|$ increases by oscillating and achieves a maximum at $d/\sigma \sim 250 - 300$; for $S_{\rm d} = 0$, the value of $\ln|Q|$ achieves a maximum at $d/\sigma \sim 40 - 50$ and then decreases by oscillating; at large thicknesses the diffraction suppression of emission occurs; for $S_{\rm d} = -0.5$, the value of $\ln|Q|$ increases, achieves a maximum already at $d/\sigma \sim 30 - 40$, and then decreases by oscillating; at large thicknesses the diffraction suppression of emission again takes place. Thus, the anomalously strong increase in the emission intensity is observed outside the DRR near the short-wavelength (long-wavelength) boundary for $S_{\rm d} = 0$ and -0.5 (for $S_{\rm d} = 0$ and 1) at small layer thicknesses, which is important for the development of miniature lasers.

The dielectric boundaries, as mentioned above, provide a weak feedback for the non-diffracting eigenmode, and the anomalously strong increase in the emission intensity takes place for this mode as well. Note, however, that this occurs at much greater CLC thicknesses (for $d/\sigma \sim 350 - 450$).

Consider now the influence of the concentration of dye molecules in a CLC layer on the increase in the emission intensity. Figure 3 presents the dependences of $\ln|Q|$ on the parameter $\ln(j\varepsilon''_{\rm m})$ for $\alpha = 1$ ($j = 1$ for $S_{\rm d} = 1$ and -0.5, $j = 2$ for $S_{\rm d} = 0$). The first row shows the dependences for the wavelength 0.613 μm outside the DRR near its short-wavelength boundary for CLC layer thicknesses presented in figures (these thicknesses were chosen so that they correspond to peaks in the dependences of $\ln|Q|$ on the layer thickness for the specified wavelength). The second row corresponds to the wavelength 0.615 μm within the DRR near its short-wavelength boundary. For the incident wave with the non-diffracting intrinsic polarisation [curves (*2*)], $\ln|Q|$ passes through a maximum with increasing $\ln(j\varepsilon''_{\rm m})$. For the incident wave with the diffracting intrinsic polarisation [curves (*1*)], emission is suppressed within the DRR, while outside the DRR the quantity $\ln|Q|$ passes through two maxima with increasing $\ln(j\varepsilon''_{\rm m})$. For $S_{\rm d} = 0$ and 1 near the short-wavelength DRR boundary, an anomalously strong increase in the emission intensity takes place at comparatively low concentrations of dye molecules and a comparatively small layer thickness. Near the long-wavelength DRR boundary, this effect is observed for $S_{\rm d} = 0$ and -0.5.

In the presence of dielectric boundaries (for $\alpha \neq 1$), a weak feedback for the non-diffracting eigenmode provided by them leads to the displacement of the maximum of the dependence $\ln|Q|$ on the parameter $\ln(j\varepsilon''_{\rm m})$ to the small values of the latter. The influence of dielectric boundaries on this dependence for the diffracting eigenmode is insignificant because the strong diffraction interaction of light with the medium suppresses the influence of dielectric boundaries (of coarse, when α close to unity).

4. Conclusions

We have analysed the properties of emission amplification in a CLC doped with laser dye molecules in the presence of the pump wave. The influence of the layer thickness, the dielectric boundaries, and the dye concentration on amplification has been studied.

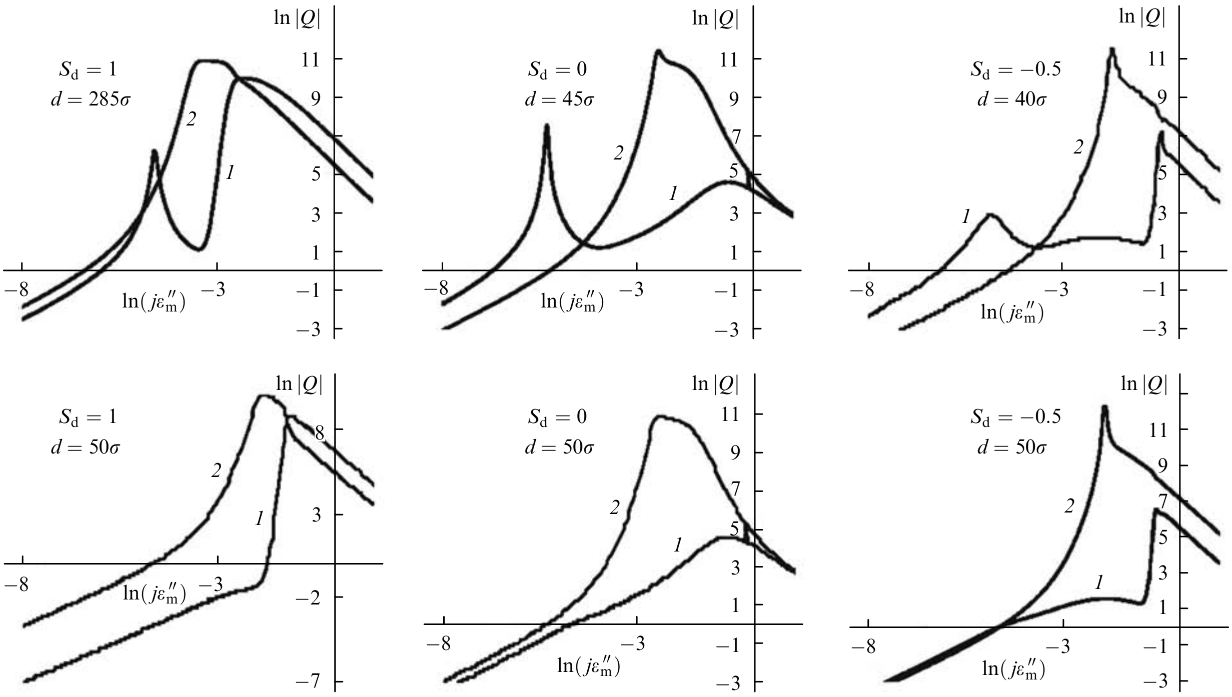

Figure 3. Dependences of the emission intensity $\ln|Q|$ on the parameter $\ln(j\varepsilon''_{\rm m})$ for the incident light wavelength 0.613 μm (the first row) and 0.615 μm (the second row) in the absence of dielectric boundaries; $j = 1$ for $S_{\rm d} = 1$ and -0.5, and $j = 2$ for $S_{\rm d} = 0$. Parameters of the CLC layer and curve numeration is as in Fig. 1.

Note that the problem of the propagation of radiation in the resonator with the active element with the constant gain $4\pi n''_{1,2}/\lambda$ does not correspond to the real process. The gain decreases with increasing the intensity of the wave propagating in the medium. This is explained by the features of the inverse state production: when the energy accumulated in the active element is very high, the rate of simulated transitions exceeds the pump rate. In this case, the population difference of the excited and ground states drastically decreases, resulting in the decrease in the gain and, hence, in the intensity saturation [27]. Because the interaction of radiation with the amplifying medium becomes nonlinear and nonstationary, the linear approximation cannot be used.

Note in conclusion that the results of this paper can be used for the development of miniature lasers with circularly polarised fundamental mode.

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