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Lasers on cholesteric liquid crystals: mode density and lasing threshold

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The density of states approach [J. Bendickson et al., Phys. Rev. E **53**, 4107 (1996)] was applied to a simple dielectric plate and a formula was found for the spectrum of the threshold gain for lasing. Then the validity of the same approach was verified for cholesteric liquid crystals (CLC) having helical structure. For non-absorbing CLC, the dependences were found of the minimum threshold gain on the layer thickness and the optical anisotropy of the material. The contribution of the dye absorption was discussed separately. The experimental data presented are in good agreement with threshold gain calculations.

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Although lasing effects in cholesteric liquid crystals (CLC) were observed many years ago [1] only recently these materials were recognized [2] as promising materials for low-threshold mirrorless dye microlasers with a distributed feedback (DFB), which can be constructed in the form of planar multi-pixel structures easily integrated with planar amplifies [3] and other optical devices. Chiral liquid crystals have natural periodicity and are considered as one-dimensional [2] or three-dimensional [4] photonic crystals. In CLC the spectral position of the bandgap is determined by period of the helix (pitch P_0) which is very sensitive to different external factors providing excellent frequency tunability of CLC lasers. In comparison to other photonic crystals, the optical anisotropy of CLC is very large, $n_{\|}-n_{\perp}\approx 0.2$ ($n_{\|}$ and $n_{\perp}$ are principal refractive indices) and this provides efficient feedback and low threshold for lasing.

The threshold conditions for lasing in conventional dye DFB lasers are calculated long ago [5] but these results cannot directly be applied to helical CLC due to circular polarization of the eigenmodes and a very particular shape of the Bragg band. Very rough estimation of the threshold pump has been made [6] using a Lorentz shape of the Bragg band quite different from the real shape. Cao et al. [7] have made an attempt at discussion of the laser threshold based on the DOS (density of optical states) technique earlier developed for non-chiral photonic band crystals [8], however, have not obtained quantitative results. The same DOS technique was applied [9] to calculation of a spontaneous emission spectrum in amplifying medium but laser threshold was not discussed. The only quantitative results on the CLC lasing threshold have been obtained by Palto [10] who simulated numerically the thickness dependence of the threshold using a software based on the precise solution of the Maxwell equations for anisotropic, periodic medium with light amplification.

Here, to get clear physical picture related to the CLC lasing threshold, the problem is considered in a very simple and systematic way, based on the DOS concept. At first, the analytic formulas for DOS and the threshold gain spectra are found for a uniform non-absorbing plate of finite thickness. Then the same formulas will be applied to a non-absorbing helical CLC. The results are compared with numerical simulation made for the exactly same parameters of CLC. Already at this point, new results will be obtained as far as the threshold dependence on the cell thickness and optical anisotropy is concerned. Then we discuss the CLC with absorption and compare the results with published and our own experimental data.

DOS= $dk/d\omega$ is inversed group velocity and defined as density of wavevectors k per unit volume and unit angular frequency ω. In a laser device, the reflections from sample boundaries or from a periodic structure creating the feedback may alternatively be taken into account *via* DOS dramatically dependent on these reflections [8]. Here, the spectrum of DOS(ω) is considered as a property of the "cold" cavity, independent of the light absorption or amplification. Both positive and negative absorption are considered at the next step as correction factors. It is possible because, at the lasing threshold, the real part of the wavevectors is much lager than the imaginary one.

For calculation of DOS we only need the complex transmission coefficient of a sample studied $t = X + iY$. Since the total phase accumulated by light wave during

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Thickness dependence of maximum DOS (ρ_{max}), minimum threshold gain coefficient (α_g^{th}) and threshold pump power (P) for DCM dye (anisotropy $\delta = 0.1245$)1

L (μm)	2	4	6	7.5	10	20	30	40	50	60
ρ_{max}	1.57	3.0	5.03	6.05	8.01	21.8	45.9	104	138	201
α_g^{th} (cm^{-1})	7600	1720	660	480	250	46	14.4	4.8	3	1.64
P (DCM) (kW/cm^2)	54000	12600	4840	3520	1830	336	105	35	22	12.1

propagating through the sample of thickness L is $\varphi = kL$ and $\tan\varphi = y(\omega)/x(\omega)$, the DOS is [8]

$$\rho(\omega) = (1/L)(Y'X - X'Y)/(X^2 + Y^2) \quad (1)$$

where primes mean derivatives with respect to ω.

a) *A transparent plate.* Now we find DOS for a simple transparent plate and then apply the result to the threshold calculation. For the normal light incidence, the complex transmission of a plate with refraction index n placed in air ($n = 1$) is [11]

$$t = \frac{t_{12}t_{23}e^{ikL}}{1 + r_{12}r_{23}e^{i2kL}} = \frac{ae^{ikL}}{1 + be^{i2kL}} = X + iY, \quad (2)$$

$t_{1,2}t_{2,3} = 4n/(n-1)^2 = a$; $r_{1,2}r_{2,3} = -[(n-1)/(n+1)]^2 = -R^2 = -b$ are the amplitudes of transmission and reflection at the two boundaries and $k = (2\pi/\lambda_0)n$. From here and Eq.(1)

$$X = \frac{a(1-b)\cos kL}{1 - 2b\cos 2kL + b^2}; \quad Y = \frac{a(1+b)\sin kL}{1 - 2b\cos 2kL + b^2}, \quad (3)$$

$$\rho_{pl}(\omega) = \frac{n}{c_o L} \cdot \frac{1 - b^2}{1 - 2b\cos 2kL + b^2}. \quad (4)$$

The $\rho_{pl}(\omega)$ function is normalized by $\rho = n/c_0$. It is a function oscillating around 1 with maxima (at $\cos 2kL = -1$) equal to

$$\rho_{max} = \frac{1-b^2}{(1-b)^2} = \frac{1+b}{1-b} = \frac{1+R^2}{1-R^2} \ \text{ or } \ R^2 = \frac{\rho_{max}-1}{\rho_{max}+1}. \quad (5)$$

The amplitude condition for the laser threshold corresponds to the exact compensation of all relevant losses (reflections R_1, R_2 and positive absorption coefficient α_a) by positive gain coefficient α_g for a round trip of photons $2L$ in the layer [12]. For $R_1 = R_2 = R = (n-1)/(n+1)$ and on account of Eq.(5), we have

$$\alpha_g - \alpha_a = -\frac{1}{L}\ln R^2 = -\frac{1}{L}\ln\frac{\rho_{max}-1}{\rho_{max}+1}. \quad (6)$$

Therefore, laser threshold condition for a dielectric plate is reformulated in terms of DOS. Formula (6) seems to be new; at least, I could not find it in literature (note the difference with an approximate expression [7] valid only for $R \approx 1$). However, it is not evident yet that Eq. (6) may be applied to calculation of lasing threshold in CLC. Therefore we should prove validity of (6).

b) *Transparent CLC.* We consider a non-absorbing, unlimited in plane x, y CLC layer of thickness L with the helical axis along z and confined from both sides by a medium whose refraction index coincides with the average refraction index of CLC. Its complex transmission coefficient is known [13, 14]:

$$t = \frac{2q_0\beta e^{iq_0L}}{2q_0\beta\cos\beta L + i(\beta^2 - k^2 + q_0^2)\sin\beta L}. \quad (7)$$

The wavevectors $\pm\beta$ of the two circularly polarized eigenwaves, which suffer Bragg diffraction upon propagating in two opposite directions along the helix axis, satisfy equation $\beta^2 = k^2 + q_0^2 - k\sqrt{4q_0^2 + k^2\delta^2}$, where $q_0 = 2\pi/P_0$ is the helix wavevector, $k^2 = (\omega/c_0)^2\langle\varepsilon\rangle$ [15]. Here, average dielectric permittivity is $\langle\varepsilon\rangle = (\varepsilon_\square + \varepsilon_\perp)/2$ and its anisotropy $\delta = (\varepsilon_\square - \varepsilon_\perp)/(\varepsilon_\square + \varepsilon_\perp)$. For simplicity we select thicknesses of the layers studied to be equal to the integer number of periods of the helix, $L = m/q_0 = 2m\pi P$. Then the real and the imaginary parts of the complex transmission of CLC are:

$$X = \frac{4q_0^2\beta^2\cos\beta L}{4q_0^2\beta^2 + k^4\delta^2\sin^2\beta L};$$

$$Y = \frac{2q_0\beta(\beta^2 - k^2 + q_0^2)\sin\beta L}{4q_0^2\beta^2 + k^4\delta^2\sin^2\beta L}. \quad (8)$$

The DOS and the threshold gain coefficient for CLC are calculated as before, see Eqs.(1) and (6). In Fig.1a the DOS spectrum *per unit length* (1 μm) of a CLC layer having $P_0 = 0.4\,\mu$m, $\delta = 0.1245$ ($n_\parallel = 1.7$, $n_\perp = 1.5$) is shown for 10 μm thick layer. The ordinate is normalized to $\langle n\rangle/c_0$ ($\langle n\rangle = \langle\varepsilon\rangle^{1/2} = 1.603$), hence $\rho \approx 1$ outside the stop-band. The $\rho(\lambda)$ spectrum has characteristic maxima located at both the short- and long-wavelength edge of the stop band. One of them is usually associated with a lasing band and determined by the gain spectrum of a dye. The dependence of DOS maximum values on cell thickness is shown in Table. It is well fit by parabola $\rho(L) \approx 1 + 0.054L^2$.

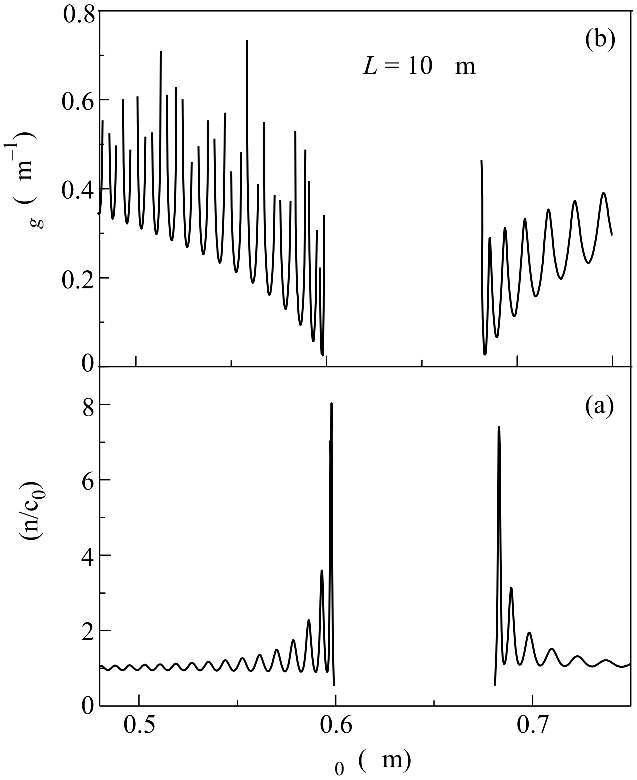

Fig.1. Spectra of DOS (a) and the threshold gain coefficient α_g (b) for a $10\,\mu$m thick non-absorbing CLC layer ($\delta = 0.1245$)

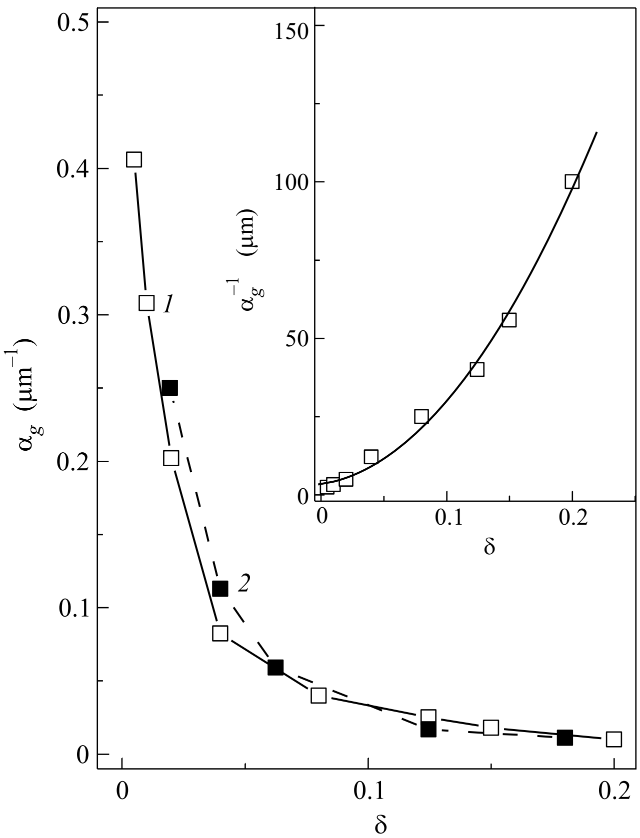

Fig.2. Dependence of the threshold gain coefficient on the CLC anisotropy δ calculated from DOS approach (curve *1*) and using software [16] (curve *2*). Inset: Inversed threshold gain coefficient vs anisotropy δ, $L = 10\,\mu$m

Using DOS and Eq.(6) we calculate the spectra of the threshold gain coefficient for different layer thickness and dielectric anisotropy over a wide range of wavelength. One of such a spectra (for $L = 10\,\mu$m and $\delta = 0.1245$) is shown in Fig.1b. Note that, for simple laser devices, the absolute minima $\alpha_g^{\min}$ of the threshold curve closest to stop-band edges are the most important. However, the whole wavelength dependence of it is also useful for more complicated (e.g. waveguiding) devices in which eigenmode spectral positions may not coincide with the minimum threshold gain. The thickness dependence of $\alpha_g^{\min}$ is shown in Table. Within a wide range of L ($2{-}60\,\mu$m) it approximately follows the law $\alpha_g^{\min} \propto L^{-3}$ surprisingly similar to that predicted by the two-coupled-wave diffraction theory for a non-chiral DFB lasers [5]. For helical CLC, a similar dependence was also found by simulating [10].

Curve *1* in Fig.2 shows the dependence of the threshold gain on the optical anisotropy within a whole range of $\delta = 0{-}0.2$ one can encounter in CLCs. The inverse of the minimum threshold is fit by the polynomial as $1/\alpha_g^{\min} = 3.5 + 58\delta + 2070\delta^2$. It follows the law $\alpha_g^{\min} \propto \delta^{-2}$ predicted in [5] only at relatively large anisotropy. Curve *2* in Fig.2 is the result of simulation made in framework of the present work using software [10, 13]. The parameters of CLC for the two curves were the same and the gain (without anisotropy) was varied to find a singularity in transmission i.e. lasing threshold. The $\pm 30\%$ difference between curves *1* and *2* may be attributed to more precise account of reflections from the boundaries in numerical simulations. From this comparison we can conclude that the relatively simple and fast calculations based on Eqs. (1), (5) and (6) indeed work for CLC.

c) *Comparison with experiment*. Table and Fig.2 predict values of the *threshold gain* for CLC without absorption in isotropic gain approximation. For example, a $50\,\mu$m thick CLC with typical $\delta = 0.1245$ has $\alpha_g^{\min} \approx 3\,\mathrm{cm}^{-1}$. Using this value we can calculate the *threshold population density number* N_e of dye molecules in the excited state and then the threshold pump power. For the latter we assume a typical case when a dye-doped CLC is pumped at $\lambda_0 = 532\,$nm by short pulses ($t_p = 10\,$ns). To find N_e we only need a *molecular cross-section for spontaneous emis-*

sion $\sigma_e(\omega)$ because, in the absence of absorption at the lasing wavelength, $\alpha_g^{\min} = N_e\sigma_e(\omega)$ [12]. The cross-section is $\alpha_e(\omega) = (2\pi c/\omega\langle n\rangle)^2 f(\omega)$ where the spectral function $f(\omega)$ is a dye luminescence spectrum at low pump energy. It should be normalized to the ratio $\Phi/8\tau$ where Φ is quantum efficiency and τ the lifetime of dye excited state: $\int_0^\infty f(\omega)d\omega = \Phi/8\tau$. The most popular laser dye DCM (Dicyanomethylene)-2-methyl-6-(4-dimethylamino-styryl)-4H-pyran) has $\Phi = 0.27$ (data of Exciton), and $\tau = 1.25\,$ns [7] and rather a wide emission spectrum well separated from the absorption band. From the luminescent spectrum measured by us we find the maximum cross-section $\sigma_e^{\max} = 0.82\cdot10^{-16}\,\text{cm}^2$ at $\lambda_{\max} = 0.587\,\mu$m. The entire spectrum $\sigma_e(\omega)$ is shown in Fig.3. Assuming that $\lambda_{\max}$ coincides with the low-frequency edge of the stop-band we find the threshold value of N_e: $N_e^{th} = \alpha_g^{\min}/\sigma_e^{\max} \approx 3.6\cdot10^{16}\,\text{cm}^{-3}$.

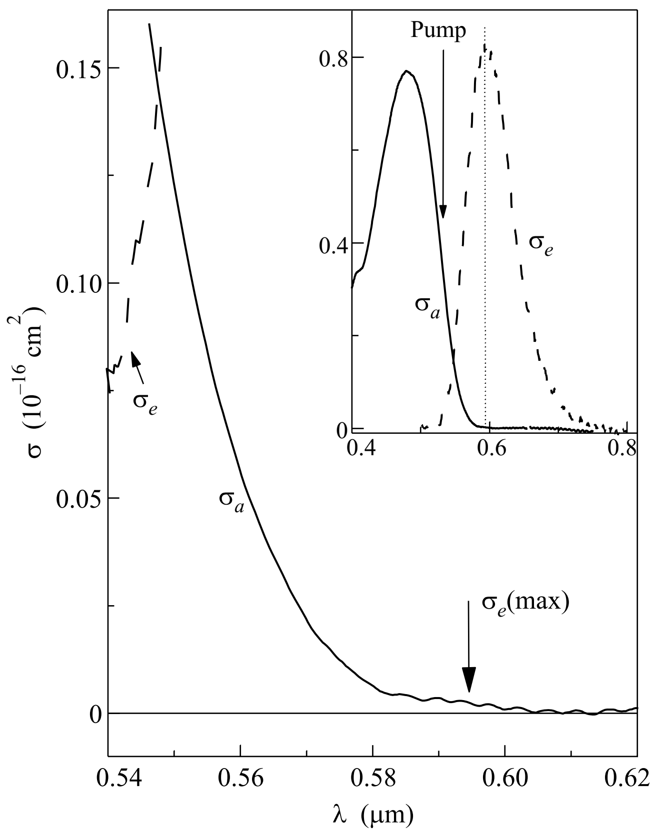

Fig.3. Inset: Experimental spectra of emission (σ_e) and absorption (σ_a) cross-sections for DCM dye molecules in units $10^{-16}\,\text{cm}^2$. The arrow shows the pump wavelength 532 nm. Main plot: same cross-sections on the same but blown scales. The arrow marked as σ_e(max) shows the wavelength of maximum of $\sigma_e(\lambda)$ seen in the Inset

Then, having in mind that a pump pulse duration $t_p \approx 10\tau$ provides the steady state regime, we find the excitation rate of dye molecules $g = N_e^{th}/\tau$. For calculation of the *threshold pump power*, we only need the spectrum of the pump absorption coefficient $\alpha_a(\omega)$ related to absorption cross section of DCM molecules and their number density in the ground state $\alpha_a(\omega) = \sigma_a(\omega)N_g$:

$$P = \frac{\hbar\omega N_e^{th}}{\alpha_a(\omega)\tau} = \frac{\hbar\omega\alpha_g^{\min}}{\sigma_e^{\max}\alpha_a(\omega_{\text{pump}})\tau}. \tag{9}$$

The spectrum of $\alpha_a(\omega) = D(\omega)/N_g L\log_{10} e$ is calculated from the measured optical density $D(\omega)$ of a DCM solution (for typical dye concentration of 0.5 wt%, $N_g = 1.2\cdot10^{19}\,\text{cm}^{-3}$) at a particular thickness $L = 27\,\mu$m in the isotropic phase of a popular nematic mixture E7. Finally, for $50\,\mu$m thick CLC, from Fig.3 we find $\sigma_a = 0.35\cdot10^{-16}\,\text{cm}^2$ at $\lambda_0 = 0.532\,\mu$m and calculate $P = 26\,\text{kW/cm}^2$ (for other thicknesses see again Table).

Up to now we considered only that term in formula (6) which is related to the resonator losses. Any *absorption loss spectrum* $\alpha_a(\omega)$ will be added to the threshold gain spectrum, shown in Fig.1b and the total spectrum will be shifted upward. Therefore, the threshold pump power could be increased dramatically. Consider the blown spectra of the two cross-sections in Fig.3. If the edge of the stop band is located at, say, $\lambda_0 = 0.61\,\mu$m slightly exceeding σ_e(max), σ_e would be reduced only by 5%. At the same λ_0, the absorption cross-section is negligible and the threshold pump would be close to the absolute minimum of $26\,\text{kW/cm}^2$. However, if for instance the stop-band edge is at $\lambda_0 = 0.57\,\mu$m, then σ_e is reduced by 25% and, what is much more important, σ_a increases up to $2\cdot10^{-14}\,\text{cm}^2$. This would add loss coefficient $\alpha_a = 24\,\text{cm}^{-1}$ to the ideal gain of $3\,\text{cm}^{-1}$ and increase pump power 9-times up to the value about $230\,\text{kW/cm}^2$. Moreover, some extra losses originate from an incorrect choice of the cell thickness because the extra material behind the illuminated layer of thickness $1/\alpha_a$ (at ω_{pump}) will not be pumped and the laser threshold will dramatically increase. In addition, some losses originate from the light scattering on the defects of the CLC structure.

In experiment, the lowest pumping power reported are by Il'chishin et al. [17] ($150\,\text{kW/cm}^2$, $L = 20{-}30\,\mu$m, Phenalenon dye), Kopp et al. [2] ($280\,\text{kW/cm}^2$, $L = 20{-}30\,\mu$m, Pyrromethene dye) and Morris et all. [18] ($1\,\text{MW/cm}^2$, $L = 7.5\,\mu$m, DCM) and also in our experiments, see below. Usually dye parameters do not differ much and we can make rough comparison with calculations. The data of Il'chishin and Kopp are close to data in Table (about $100{-}300\,\text{kW/cm}^2$). As to the Morris data obtained for

DCM dye, it is even smaller than the calculated limit ($3.5\,\mathrm{MW/cm^2}$) that can be attributed either to some inaccuracy in the pump spot diameter ($\sim 100\,\mu$m, specified approximately) or to our theoretical overestimating of the threshold due to isotropic gain approximation. Numerical simulation predicts that on account of anisotropy the threshold should be about 2–3 times lower.

Our results for the threshold measurements on the two CLC samples are shown in Fig.4. In both panels

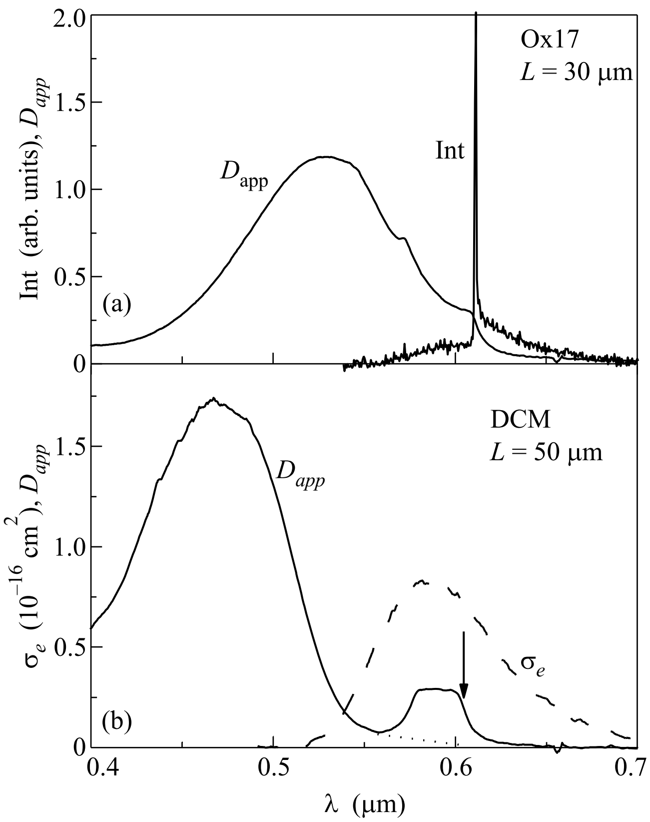

Fig.4. Apparent optical density D_{app} with the photonic stop-band seen and emission intensity of two different CLC samples. (a) 30 μm thick CLC doped with Oxazine-17 dye; intensity of lasing (Int) at the long-wavelength band edge superimposed on the absorption spectrum. (b) 50 μm thick CLC doped with DCM dye with the laser generation line (shown by the arrow) in the spectral zone outside of the absorption band; σ_e is the emission cross-section spectrum of DCM in this CLC sample

(a) and (b), the apparent optical density D_{app} (without correction for the Bragg diffraction) are shown by solid lines. For the 30 μm thick CLC cell, see Fig.1a, the Bragg band is located between 0.572 and 0.614 μm and considerably overlapped by the absorption spectrum of dye (in this case, Oxazine-17). The dye luminescence is observed at $\lambda = 0.56-0.70\,\mu$m and intense lasing line is seen at $\lambda = 0.612\,\mu$m. The lasing threshold is found at $460 \pm 150\,\mathrm{kW/cm^2}$ (pump pulse energy $1.3\,\mu$J, $t_p = 7$ ns, pumping area $S = (5 \pm 2) \cdot 10^{-4}\,\mathrm{cm^2}$ measured with a blade translated by a microscopic screw) that is about 5 times *higher* than the theoretical value for $L = 30\,\mu$m. The second sample is 50 μm thick layer of a CLC doped with DCM dye with a laser generation line in the spectral zone outside of the absorption curve (shown by an arrow in Fig.4b. In this case, the threshold energy is 40 ± 5 nJ/pulse, t_p and S are the same and the threshold power is $14 \pm 7\,\mathrm{kW/cm^2}$ that is 2 times *lower* than our theoretical value of $26\,\mathrm{kW/cm^2}$ for $L = 50\,\mu$m (see the Table). Since the two laser dyes have similar properties, such a large difference between the first and second samples (almost an order of magnitude) is attributed to the different location of the stop-band with respect to the absorption spectrum.

In conclusion, the analytic technique for DOS calculation [8] has been applied to a simple dielectric plate and Eq.(6) was found to be very useful for calculation of the spectra of threshold gain in lasing helical CLC. For non-absorbing CLC, the dependences of the minimum threshold gain on the layer thickness and the optical anisotropy of the material were calculated. The spectrum of absorption was considered as an additional contribution that just shifted upward the threshold gain spectrum. The threshold calculation were compared with the experimental data and the results of the comparison allows one to use the suggested simple and fast technique for routine estimation of the threshold of CLC lasers.

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