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Lasing in a hybrid aligned cholesteric

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Abstract. Optically pumped laser generation at the long-wavelength of cholesteric photonic band gap is registered in a dye-doped cholesteric cell with hybrid alignment, where long axes of the cholesteric molecules are parallel to the substrate at one substrate, but perpendicular near the opposite substrate. Hybrid director alignment in the cell is confirmed by polarization optical microscopy observations in reflection and transmission modes.

1. Introduction

Lasing in a dye-doped cholesteric (Ch) liquid crystal (LC) usually is obtained in a cell with uniformly planar (parallel to the substrates) Ch layers. Due to the phenomenon of selective light reflection at appropriate relations between the Ch pitch and light wavelength, such planar Ch layers form a resonator for circularly polarized light waves of the same handedness as that of the Ch chirality [1-4]. Quality of the planar alignment is therefore one of the key factors for lasing [5] in cholesterics (Ch-s). In thick cells good enough planar alignment is hard to achieve. For this reason thin cells of about $5-15\mu\text{m}$ are usually used for lasing. Recently we have shown [6,7] that laser generation can be obtained also in thick imperfectly aligned cells with a large number of defects. In this study, we check whether lasing is possible in a dye-doped Ch cell with hybrid surface alignment, where long axes of the Ch molecules are parallel to the substrate at one substrate, but perpendicular (homeotropic alignment) near the opposite substrate. Thereby Ch layers are expected to be strongly curved through the cell thickness. However, there is no evidence that the cell with substrates providing hybrid alignment for nematics will do the same effect for a short-pitch Ch. Short-pitch Ch-s show specific elasticity and surface anchoring properties in comparison with their long-pitch and untwisted (nematic) counterparts [8].

Identification of hybrid alignment is a complicate issue for nematics [9]. The problem is that the polarization optical microscopy (POM) textures of a hybrid nematic cell are qualitatively not distinguishable from those for cells with uniformly tilted or even planar director. Special techniques such as Fluorescent Polarization Confocal Microscopy [10,11], PolScope [12,13] or Magnetic Null technique [9,14-16] have to be applied to detect hybridity in a nematic cell. For Ch-s, especially with the short pitch, it still remains to be an open problem. We demonstrate that a hybrid-aligned short-pitch Ch cell shows

distinctive POM textures in transmission and reflection modes, which allow for unambiguous identification of its hybrid alignment.

At first glance a Ch cell with hybrid alignment should not show lasing, because distortion of the Ch layers through the cell thickness would destroy the resonator. Surprisingly, for a hybrid cell we register lasing picks at the edge of the Ch Photonic Band Gap (PhBG) similar to those observed in planar Ch cells. POM textures and experimental registration of lasing in the hybrid Ch cell reported in this paper suggest that at the substrate with planar alignment the layers remain planar for some finite microscopic distance from the substrate.

2. Experimental

We prepared a cell assembled of two substrates. One of them, coated with the polymer SE5661, provides homeotropic orientation of the molecules at its surface, whereas the other one, coated with polymer PI2555 aligns nematic molecules parallel to its surface. The gap between the substrates was fixed with spherical glass spacers of calibrated diameter, added into the UV-sensitive glue, applied between the substrates along their edges. After photo-polymerization of the glue with a UV-lamp, the cell thickness, measured with the light interference technique from transmission spectrum data, was calculated to be $d = 16.6\mu\text{m}$.

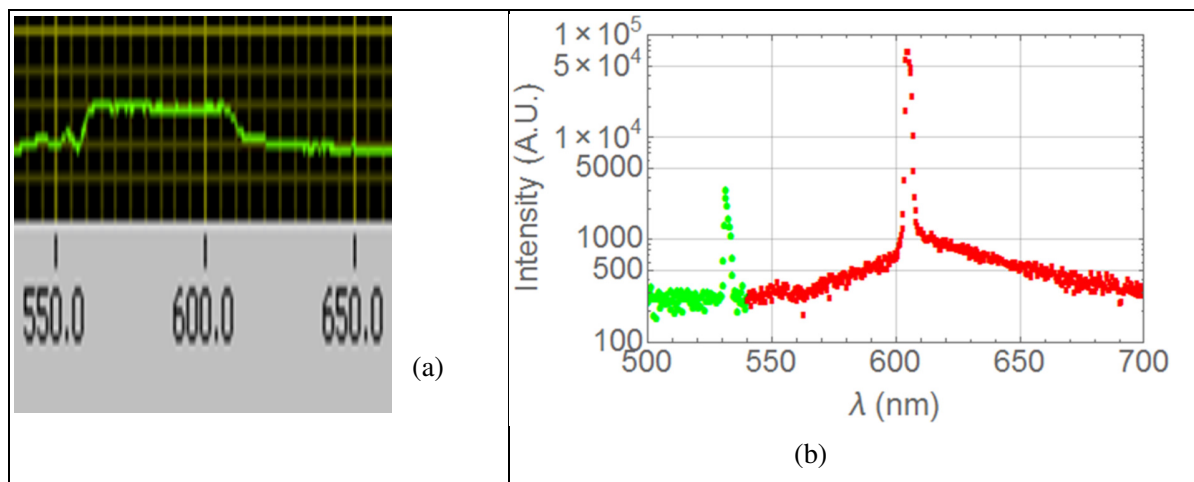


Fig. 1. (a) Reflection spectrum of the planar oriented cholesteric cell filled with a mixture HNG751600-100 + 24% S811. On the horizontal axis the light wavelengths is given in nm; the vertical axis corresponds to the intensity of the reflected light in arbitrary units; (b) Emission spectra of the cholesteric planar cell (HNG751600-100 + 24% S811 + 0.5% NR). Lasing line is observed at $\lambda = 605\text{nm}$ (red dots), which falls on the long-wavelength edge of the cholesteric PhBG. The pick at $\lambda = 532\text{nm}$ (green dots) corresponds to the excitation laser light.

The gap of the cell was filled with a mixture of the nematic HNG751600-100 mixed with 24 wt.% of chiral agent S811 and 0.5 wt.% of the fluorescent dye Nile Red. The spectral band of selective reflection for the cholesteric without dye in a planar cell, measured with a fiber micro-spectrometer Ocean Optics USB 2000, is between 560 and 605nm (Fig.1a). For a Ch, which is a 1D photonic crystal, the position of the selective reflection band visualizes the position of the Ch PhBG.

Composition of the cholesteric mixture was chosen such that the maximum of the fluorescence emission band (FEB) for the NR dye in the LC matrix falls on the long-wavelength edge of the Ch PhBG. The FEB maximum under excitation with green ($\lambda = 532\text{nm}$) light for the NR dye, dispersed in the LC matrix, is at 610nm. As expected, laser generation in a Ch planar cell with the same composition (HNG751600-100 +

24% S811 + 0.5% NR) was registered at the long-wavelength edge of the Ch PhBG (Fig. 1b). For pumping of laser generation we use a pulse laser with the wavelength $\lambda = 532nm$, the pulse width of 1 ns, tunable ($1Hz \div 20kHz$) repetition frequency (chosen to be of $150Hz$) and pulse energy up to $2\mu J / pulse$.

3. Results and discussion

3.1. Identification of hybrid alignment

POM observations in the transmission mode show that the texture of the cell (Fig. 2) resembles a typical Ch texture, indicating planar Ch layers (helical axis perpendicular to the substrates), the so-called Grandjean texture with clearly visible oily streaks (thick lines) on the background of a set of thin lines, which resemble a "fingerprint texture", a typical texture indicating homeotropic cholesteric layers (helical axis parallel to the substrates). For the reason explained below, we call it the "pseudo-fingerprint" texture. Moving the focus of the microscope up and down, one can see that these two textures are located at different levels in the cell. The Grandjean texture looks to be located near the substrate that requires planar orientation of the molecules on its surface, whereas "pseudo-fingerprint texture" seems to be formed near the substrate with the homeotropic anchoring conditions. Homogeneous purple color of the texture in Fig. 2 is a result of selective reflection. The combination of these two textures (Grandjean texture on the background of "pseudo-fingerprint" texture) is an indirect evidence for hybrid surface alignment. We find that direct evidence can be obtained from POM observations in reflection mode. In the reflection mode the illumination of the sample is performed through the objective of the microscope in such a way, that the light, reflected from the sample, returns back into the objective. At such an observation mode the POM texture is formed by the distribution of the director in the vicinity of the upper

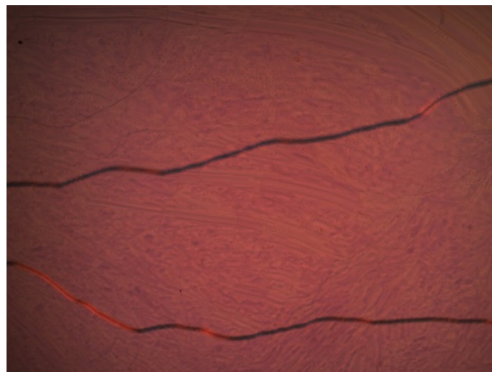


Fig. 2. POM texture of the hybrid cell between crossed polarizers in transmission mode. Thick lines are oily streaks, thin lines resemble a "fingerprint".

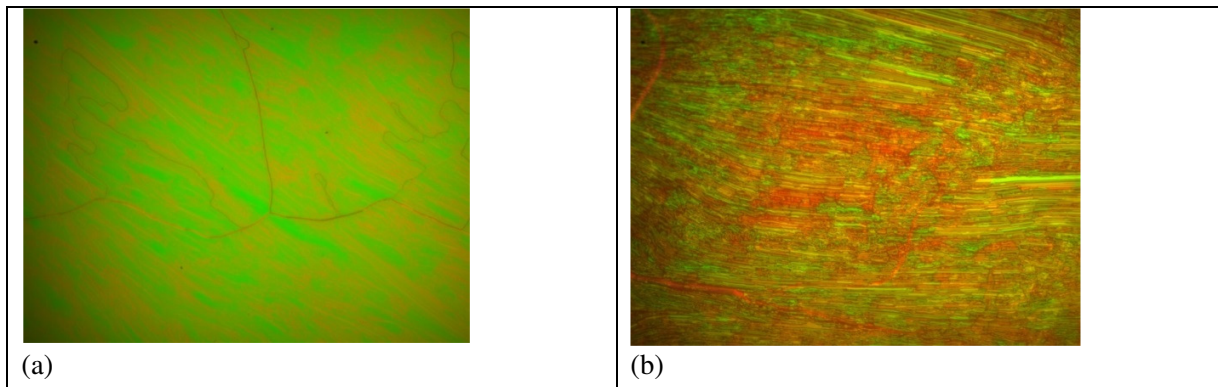


Fig. 3. POM texture of the hybrid cell observed in the reflection mode, when the light falls: on and reflects from the surface of the substrate with (a) planar and (b) homeotropic boundary conditions for the liquid crystal molecules.

substrate faced to the objective, which implies that the corresponding textures (Grandjean and "pseudo-fingerprint" textures) can be observed separately for each of the two surface - not superimposed, as it is in the transmission mode.

In Fig.3a we show the texture, obtained in reflection mode such that the cell placed on the microscope stage is faced to the objective by its substrate with planar anchoring conditions. The observed texture (Fig. 3a) is a typical Grandjean texture. Usually Grandjean texture is observed for flat cells with planar cholesteric layers. Clearly visible color of selective reflection varies from green to orange. The "psedo-fingerprint" texture is not observed, but it becomes clearly visible, if one turns the cell by its homeotropic substrate towards the objective (Fig. 3b). These two textures bring the evidence that the cell under study is a hybrid cell with the planar alignment at one substrate, visualized by the Grandjean texture and the homeotropic molecular alignment at the opposite substrate, visualized by the "pseudo-fingerprint" texture.

It should be noticed that for homeotropic cells of long-pitch cholesterics the periodicity of the "fingerprint" texture is of the order of half-pitch or a bit larger if the layers are tilted with respect to the substrates. Domains with different periodicities can coexist in the same cell, but periods in them differ usually not more than twice. The quasi-periodical set of lines seen on the "pseudo-fingerprint" texture in Fig. 3b is of the periodicity much larger ($\sim 4\mu\text{m}$) than the Ch pitch. As we suggest below its origin is analogic to the oily-streaks and Focal Conic Domain (FCD) texture, observed in thin hybrid aligned smectic cells. Therefore the "pseudo-fingerprint" texture is only apparently similar to the fingerprint texture in cholesterics. By its origin it is a typical texture of layered liquid crystals such as smectics. For this reason, as it was announced above, we call the corresponding texture observed in hybrid aligned cholesterics the "pseudo-fingerprint" texture.

The Ch pitch can be estimated from the color of selective reflection, which roughly corresponds to the light wavelength $\lambda_s \sim 0.57\mu\text{m}$. For the studied cholesteric mixture the average refractive index can be taken approximately $\bar{n} \sim 1.6$ and, thus, the pitch can be estimated as $P \sim \lambda_s / \bar{n} \approx 0.36\mu\text{m}$. Therefore the pitch and consequently the half-pitch $P/2 \sim 0.18\mu\text{m}$ appear to be significantly shorter than the typical periodicity $\sim 4\mu\text{m}$ observed for the lines in Fig.3b, which in its turn is much smaller than the cell thickness $d = 16.6\mu\text{m}$.

A set of parallel lines perpendicular to the helix direction with the periodicity of several pitches was documented in Ref. [17] for a short pitch Ch cell with substrates treated for homeotropic molecular alignment; the texture was identified as the so-called cholesteric FCD texture. Inspection of the quasi-periodical texture shown in Fig. 3b shows that the periodical lines are broken by closely packed circular domains reminiscent to FCDs formed by curved equidistant Ch layers. Then the texture in Fig. 3b can be identified as a set of oily streaks, which in some places split into FCDs. The latter is in full analogy with a similar texture in hybrid aligned smectic A cells [18]. Indeed, short-pitch Ch-s elastically are similar to smectics and because of this property, the FCD texture is one of typical Ch textures [8]. In smectics the diameter of the FCDs and the periodicity of smectic oily streaks are of the order of the sample thickness, being much larger than the periodicity of smectic layers, which is of the order of the molecular length. In the same vein, in hybrid aligned short pitch-cholesterics the size D of cholesteric FCDs [19] and the period of Ch oily streaks are defined by the thickness of the region in the cell, where layers are curved. It follows

from Fig. 3b that $D \sim 4\mu m$, which is smaller than the thickness of the sample $d = 16.6\mu m$. Therefore, we are led to conclude that the Ch layers are strongly curved in the space domain of the thickness $\sim 4\mu m$. POM texture in reflection mode from the substrate coated with the polymer PI2555 and the registration of lasing in the studied hybrid aligned Ch cell suggest (see next section) that the remaining space of the cell is filled by parallel Ch layers. We will continue discussion of this issue in Sec. 3.2

3.2. *Optically pumped emission spectra of the hybrid Ch cell*

The experiment shows that the hybrid Ch cell shows laser generation similar to that from a planar Ch cell, for which the laser peak ($\lambda = 605nm$) is recorded at the long wavelength edge of the Ch PhBG (Fig. 1a,b). However for the hybrid cell filled with the same Ch mixture instead of a single lasing line one observes several peaks closely grouped around the long-wave length edge of the Ch PhBG. The number and height of peaks vary in time.

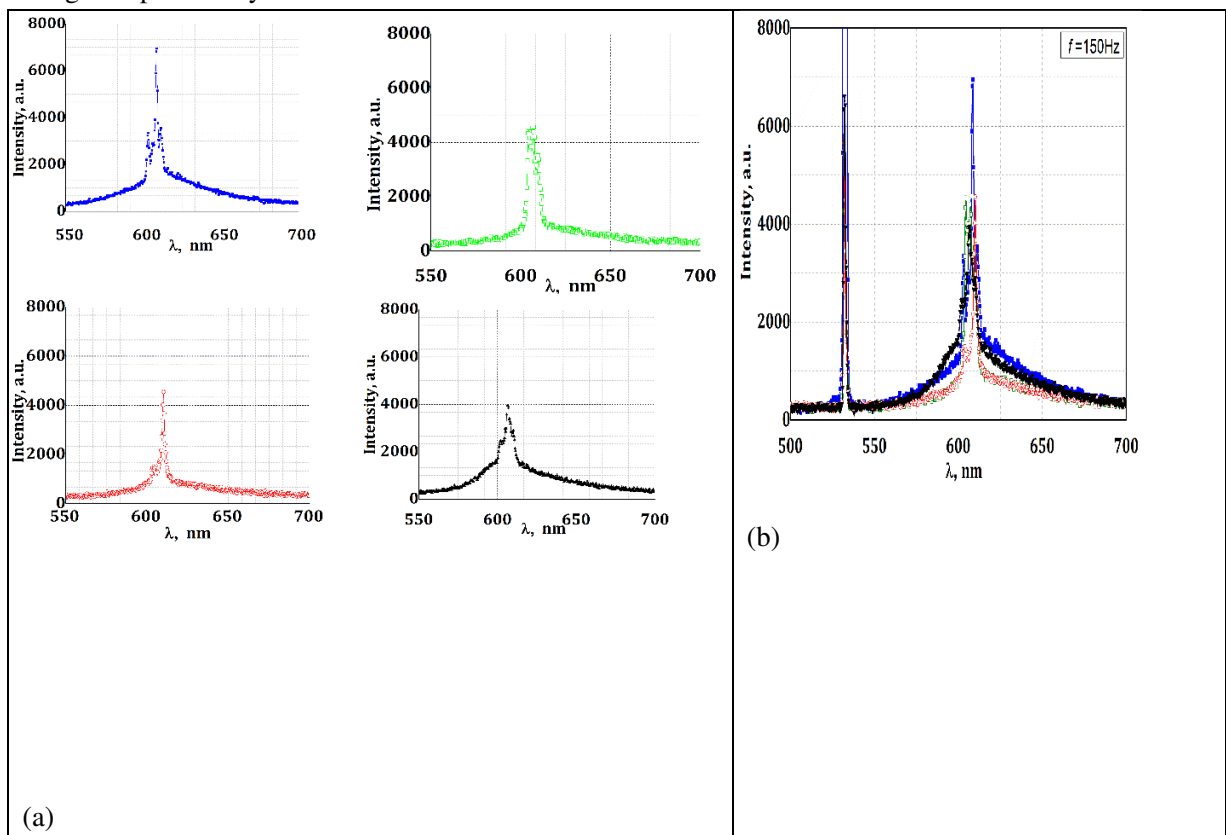


Fig. 4. Emission spectra recorded at different time moments from the cholesteric hybrid cell at the repetition frequency $f = 150Hz$ of the excitation pulse, shown (a) separately and (b) superimposed. The peak on the left at $\lambda = 532nm$ is from the excitation laser.

Emission spectra of the Ch hybrid cell with a repetition frequency $f = 150Hz$ of the excitation pulse captured at different time moments are shown separately in Fig. 4 (a) and being superimposed in Fig. 4 (b). Location of the emission peaks (Fig. 4) at the edge of the cholesteric PhBG (Fig. 1a) suggests that these peaks are of the same nature as the laser line for the planar Ch cell (Fig. 1b). The spectral width of the region (600-610nm), in which one observes the lasing peaks for the hybrid cell is of the order of 10 nm. Laser lines are observed on both sides of the wavelength $\lambda = 605nm$ corresponding to the long-wavelength edge of the PhBG of the planar cell.

The latter suggests that the laser generation takes place in the Ch layers near the substrate coated with the polymer PI2555, where the layers remain roughly parallel to the substrate, though might be significantly stretched because of their strong distortion in the bulk. It was shown theoretically and proven experimentally that stable lasing is possible only if the thickness of the planar Ch layers is at least of ten cholesteric pitches [20]. For the estimation $P \sim 0.36\mu m$ alluded to above, the thickness of the domain with planar cholesteric layers is at least $4\mu m$ (at least $\frac{1}{4}$ of the cell thickness) or can be larger.

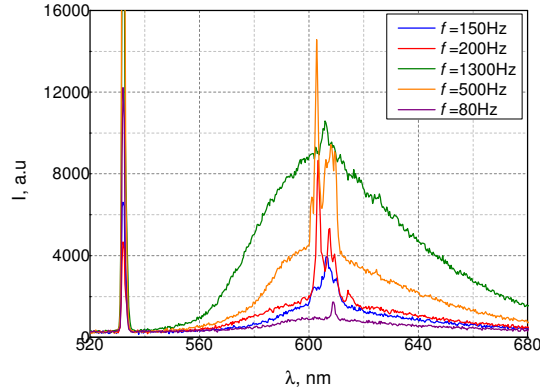


Fig. 5. Emission spectra of the cholesteric hybrid cells at excitation with the different repetition frequency of the pulse.

At the end of Sec. 3.1 we have concluded that the thickness of the sample region, where the cholesteric layers are essentially curved is of the order of $D \sim 4\mu m \ll d = 16.6\mu m$. The latter is in agreement with the above drawn conclusion that the cholesteric layers remain parallel to the substrate coated with PI255 for a finite microscope distance, which can be estimated as $\Delta d = d - D \sim 12\mu m$.

In comparison with long-pitch cholesterics or their untwisted nematic counterparts, where the orientation of the director varies rather smoothly, almost linearly, from one substrate to the other, the elasticity and surface anchoring of short-pitch cholesterics is more complicate. At hybrid alignment the director field suffers bend deformation near the substrate with the homeotropic anchoring but splay deformation near the substrate with planar alignment. Energetically bend director deformation in cholesterics is comparable to that in nematics, whereas because of the Ch orientational periodicity, the splay deformation costs high elastic energy [8]. Considerable amount of theoretical and experimental work was done to establish the organization of the Ch layers for a long-pitch Ch in homeotropic and hybrid cells [21-25]. In this respect short-pitch cholesterics are less studied. Polarization microscopy textures and experimental observation of lasing in the hybrid Ch cell reported in this paper suggest that at the substrate with planar alignment the layers remain planar for some finite microscopic distance from the substrate, which we estimate to be approximately $\frac{3}{4}$ of the cell thickness.

The dilatation of the parallel Ch layers near the substrate coated with the polymer PI2555 should lead to local variations of the Ch pitch along the substrate in comparison to the pitch equilibrium value and thereby can show up by lasing with different wavelengths around the Ch PhBG edge.

Redistribution of the emission power between the individual peaks, which is observed in time, indicates that the laser modes, corresponding to different lines are highly coupled. As it is suggested above, most probably, multiple lasing lines are result of the variation of the Ch pitch. Strong interaction of the laser modes indicates that spatial zones with different pitch are located in close proximity of each other across the light spot ($\sim 500\mu m$ in diameter in our experiment) of the excitation beam. In its turn the latter implies high density of edge dislocations in the sample. The situation resembles random lasing in a strongly

inhomogeneous scattering medium where different modes are also localized closely to each other such that separate modes appear to be coupled and strongly interact with each other [26,27].

We have performed a study of the dependence of emission spectra of the hybrid cell on the repetition frequency of the excitation signal (Fig. 5). At relatively low repetition frequency $f = 80\text{Hz}$, there is only one laser line (purple curve in Fig. 5) of relatively low intensity. Low intensity of the line is because of the low value of the exciting pulse energy at low frequencies. For higher frequencies (read: higher excitation intensities) the number of lasing modes and the height of their corresponding peaks increase. Dependence of the number of lasing modes on the excitation power indicates that different modes have different excitation power threshold. The lowest lasing threshold corresponds to the excitation with the frequency of $f = 80\text{Hz}$ and is registered for a line at $\lambda = 605\text{nm}$, which is the same as that registered for the planar Ch cell (Fig.1b). The intensity of the lasing lines increases with the increase of the frequency approximately up to $f = 500\text{Hz}$. At further increase of the frequencies one observes significant increase of the spontaneous emission such that its level becomes comparable to the lasing intensity. As a result the main light power transfers to the non-resonant emission; consequently the lasing lines are suppressed and cannot be resolved on the background of the spectral band from spontaneous emission as it is seen on the emission spectrum corresponding to the frequency $f = 1300\text{Hz}$ in Fig. 5.

Conclusions

Our experiments show that in the dye-doped cholesteric the laser generation is possible not only in the cells of high quality planar orientation of the cholesteric layers or low quality planar orientation in the presence of a large number of defects, but also in the hybrid cell where the orientation of the Ch layers changes from planar at one substrate to homeotropic at the opposite substrate. Instead of one laser line as it is for a planar cell, in the hybrid cell one observes multiple laser lines on both sides of the long-wavelength edge of the PhBG registered for a planar cholesteric cell. The emitted energy redistributes between these lines in time. Increasing of the repetition frequency of the excitation signal leads to the increased height and number of laser lines as well as to the increased intensity of non-resonant radiation which consumes energy and thus inhibits the growth of laser modes at high repetition frequencies of the excitation signal. At frequencies higher than 1500Hz the height of laser lines is comparable to the level of background non-resonant modes; as a result the laser lines cannot be resolved in the emission spectrum.

Another problem solved in this study is the identification of hybrid alignment in a short-pitch Ch cell. We demonstrate that a short-pitch hybrid aligned Ch cell displays distinctive textures in reflection modes. Namely, the Grandjean texture is observed for the substrate with planar alignment conditions and the “pseudo-fingerprint” texture which is characteristic of the homeotropic anchoring conditions. The latter is an analog of the oily streaks and FCD textures observed for hybrid aligned smectics. Curved Ch layers coexist with planar layers in the cell, such that the latter occupies of about $\frac{3}{4}$ of the cell thickness. Presence of such a thick domain with planar cholesteric layers is supported by the registration of the lasing at the edge of Ch PhBG.

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