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Lasing in Three Layer Systems Consisting of Cholesteric Liquid Crystals and Dye Solution

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We have investigated defect mode lasing in a multilayer system consisting of a dye doped isotropic solvent sandwiched between two cholesteric liquid crystal (CLC) cells. In this case we can use dyes not soluble in liquid crystals and avoid the degradation of the CLC structure caused by the absorption of the pumping energy. When the CLC pitches of both cells are equal, the dye emission generates the typical multi-mode lasing peaks inside the photonic stop band. In CLC mixtures whose pitches were shifted one respect to the other in such a way that only the edges of band gaps of the CLC layers overlapped, we have observed single mode lasing. The divergence of the laser beam spot is considerably lower than the one observed in dye doped CLC.

Keywords: cholesteric liquid crystal; lasing; photonic band gap

INTRODUCTION

Cholesteric liquid crystals (CLCs) possess several unique properties: periodic structure (the period can be set in a wide range from 100 nm up to infinity), 100% selective reflection of circularly polarized light and the ability to change their selective reflection wavelength changing external or internal factors (electric, magnetic and acoustic fields, temperature, local order, ...) [1]. In CLCs the period of helicoidal structure is equal to half the pitch P, and for light propagating along the helical axes, $P = \lambda_0/\mathbf{n}$, where λ_0 is the wavelength of the

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maximum reflection or the middle of selective reflection band (depending on the shape of the band) and **n** is the average refractive index. The full width at half maximum of the selective reflection band ($\Delta\lambda$) is equal to $P\Delta\mathbf{n}$, where $\Delta\mathbf{n}$ is the birefringence of a nematic layer perpendicular to the helix axis.

Presence of the helically distributed birefringence makes CLCs of practical interest for various laser applications. The use of a CLC layer as a mirror in conventional lasers was carried out long time ago. The temperature dependence of the selective reflection band was exploited to tune a broad band dye laser [2] and for optimizing the outcoupling efficiency of solid state lasers [3]. A CLC mirror was used as a laser end mirror in a solid state laser for providing mode operations [4].

There is a class of mirrorless dye lasers with optical feedback distributed throughout the gain medium [5]. When the CLC consists of luminescent molecules or it contains a luminescent dopant, it could be used as an active media. It is important for development of compact lasers that the major functional elements of a mirror-less laser are combined in one cell: active medium, cavity, and tunable selector. The existence of the selective reflection band and the ability to change smoothly the selective reflection wavelength over a wide range under the action of applied external forces make it possible to design broad-band tunable lasers based on dye-doped cholesteric liquid crystals (DD CLC). The patent of tunable lasing in CLCs was presented in [6]. The theoretical analysis was performed in [7]. For the first time lasing with a distributed feedback in a DD CLC was observed in [8].

More recently, the CLC was treated as a medium with a photonic band gap [9]. This approach allowed to explain the observed laser emission at the edge of the selective reflection band in the DD CLC. By analogy with the band gap in semiconductors, the photonic band gap can arise in the spectrum of the propagation of electromagnetic modes in periodic structures [10]. Within the band gap, the wave exponentially decays when propagating deep into the crystal and, correspondingly, the density of states in the band gap becomes considerably lower. Since the degree of spontaneous emission according to the Fermi law is proportional to the number of photon states, the spontaneous emission is suppressed within the band gap and, accordingly, increases at the band edges. A comprehensive analysis of the CLC as a photonic crystal is given in the review [11].

Moreover the consideration of the selective reflection band as a photonic band-gap stimulated the investigation of lasing in several chiral materials, such as thermotropic [12] and lyotropic [13] CLCs, blue [14] and TGB phases [15], chiral smectic C liquid crystals [16], polymer networks [17], elastomers [18] and glass forming CLCs [19].

Tunability of lasing in these systems was achieved varying the temperature[8], applying a mechanical stress [18] or an electric field [20], using photo-transformation effects [21] and by assembling cells with pitch gradient and with spatial distribution of different dyes [22,23].

Several attempts were made to optimize the lasing conditions and performance characteristics of DD CLC. It was shown that doping a CLC with a polymeric dye improves the order parameter and lowers the lasing threshold [24]. The dependence of the lasing threshold on dye concentration and sample thickness was studied in [25]. A remarkable change of lasing characteristics, obtained increasing the polymer concentration in polymer dispersed liquid crystals (PDLCs) was observed in [26]. Investigation of the influence of photo polymerization on lasing in CLC was made in [27]. Also it was found that the emission efficiency depends on temperature [28]. The low lasing threshold allows to develop a laser cascade consisting of two DD CLC cells with two different dyes, where the emission band of the first cell overlaps the absorption band of the second cell [29]. LEDs-controlled and reversible smooth tuning of lasing in DD CLC was obtained in [30,31].

The introduction of defects enhanced fluorescence and laser emission in a DD CLC system [32]. In this case lasing was observed inside the band gap. The enhancement in lasing efficiency and the reduction of pumping energy in photonic crystals with defects was predicted in [10]. The introduction of a defect into the CLC could be achieved in two ways: by replacing a part of the host medium with a material that has a different dielectric constant e.g. two layers of CLC sandwich a thin layer of an isotropic medium [33]; by introducing a phase jump inside the CLC cell [34]. The introduction of a defect and the tuning of photonic defect modes in the CLC, by means of the local deformation of the helix in the middle of the CLC layer, were considered theoretically in [35]. Recently an enhancement of lasing efficiency was observed in a dye doped CLC laser using a CLC reflector [36,37]. A significant degradation reduction of both, planar CLC structure and luminescent dye and as a result the stability of DD-CLC laser emission up to two hours and more was achieved by rotating the cell [38].

Experimentally the defect mode emission was investigated mostly in polymeric CLCs. Defect mode lasing in a dye doped cholesteric polymeric network, where the defect was done by a phase jump of the cholesteric helix at the interface of two stacked layers of the polymer film, was observed in [39]. The twist defect mode lasing in the middle of the 1-D photonic band gap has been experimentally demonstrated for a

composite film consisting of two photopolymerized cholesteric liquid crystal layers [40]. Defect mode lasing was investigated in three layered structures where between two CLC layers with the same pitch and handedness a dye doped nematic LC [41] or a dye doped CLC [42] was sandwiched. Also a defect mode lasing was studied when between dielectric multilayers a dye doped CLC [43] or a dye doped nematic LC [44] was sandwiched. In the first case, the tuning of lasing wavelengths was achieved by a change of the pitch induced by temperature, while, in the second case, it was achieved applying an electric field to induce a change of the refractive index in the layer. In the case of dielectric multilayers, to solve the problem of dye solubility in the LC materials, a defect can be created between two separated layers: for instance a nematic LC film and a polymer film doped by a dye [45]. It should be noted that the lasing threshold observed with the three-layered helical CLC is lower than for conventional DD CLC lasers [43,46].

Finally a wedge-shaped cell of dye doped glycerol, sandwiched between two polymeric CLC films to amplify the spontaneous emission, was investigated in [47].

EXPERIMENT AND RESULTS

In this paper we investigate the defect mode lasing in a three layer system consisting of a dye doped isotropic solvent sandwiched between two CLC cells. The separation of the CLC and the active medium allows: (1) to avoid the degradation of the CLC structure caused by the absorption of the pumping energy, (2) to use dyes not soluble in LCs, (3) to use the optimal thickness both for the CLC layer and for the dye solution layer (thicker dye layer and thinner CLC layer).

We used a nematic MLC-6816 (Merck, cyclohexylcyclohexanes), transparent in UV range. A right handed chiral dopant MLC-6248 (Merck) was added to the nematic in order to induce the cholesteric structure. Rhodamine-6G was used as a dye not soluble in LCs, and glycerol was used as an isotropic solvent.

The cells, consisting of the dye/solvent layer sandwiched between two CLC layers, represented a combination of four 0.8 mm thick glass plates separated by teflon spacers setting the thicknesses of each layer. The thickness of the CLC layers was 10 microns, and the thickness of the dye solution was 200 microns. In Figure 1 the scheme of three layer structure is shown.

The second harmonic of a Q-switched Nd:YAG laser (Continuum, Surelite II) was used as a pumping light source. The pulse wavelength, width, and repetition rate were 532 nm, 4 ns, and 1 Hz, respectively.

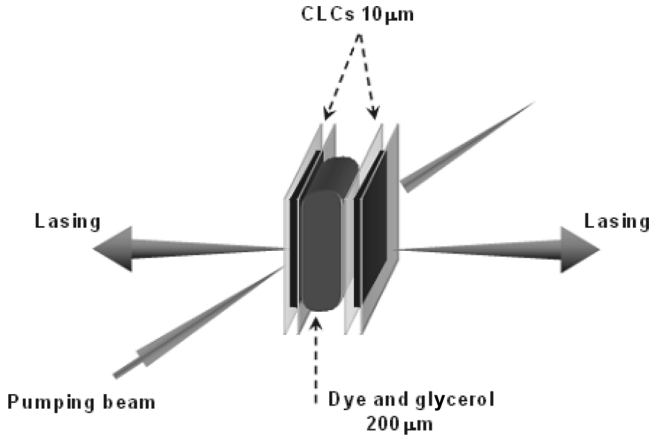

FIGURE 1 Scheme of the sandwich cell.

The laser beam was attenuated and focused by a lens ($f = 20\,cm$) to reduce the spot of the laser beam on the cholesteric cell to a few hundreds of micrometers. The pump beam irradiated the sample at an angle of 45° with respect to the cell normal. An optical fiber, coupled to a spectrometer Avantes (AvaSpec-2048, with a resolution of 0.8 nm), collected the light emitted from the sample. To get a good planar orientation of the CLC layers, the corresponding surfaces were coated with rubbed PVA (Polyvinyl alcohol) orienting layers.

At the beginning we investigated a three layered structures with CLC layers possessing equal pitches. In this case typical defect mode lasing was expected. Indeed, as shown in Figure 2 multimode lasing inside the stop band with several emission peaks was observed.

To achieve single-mode lasing the CLC cells were filled with two distinct cholesteric mixtures, whose pitches were shifted in such a way that only the edges of the band gaps overlapped. In Figure 3 the transmission spectra of each CLC layer, the spectra of dye solution emission and lasing in this cell are shown. A single mode lasing occurs in the overlapping part of CLC band gaps. In this experiment the CLC pitches were chosen to set the lasing peak near the maximum of the dye luminescence peak. In contrast with the conventional dye lasers with usual dielectric mirrors, in this kind of CLC lasers the wavelength of lasing is strongly connected with the CLC pitches. By choosing other CLC pitches, one obtains lasing at another wavelength. In Figure 4 the selective transmission of two different CLC layers superposed one to the other is shown: lasing occurs in the middle of the total band gap. The threshold of laser generation was estimated to be

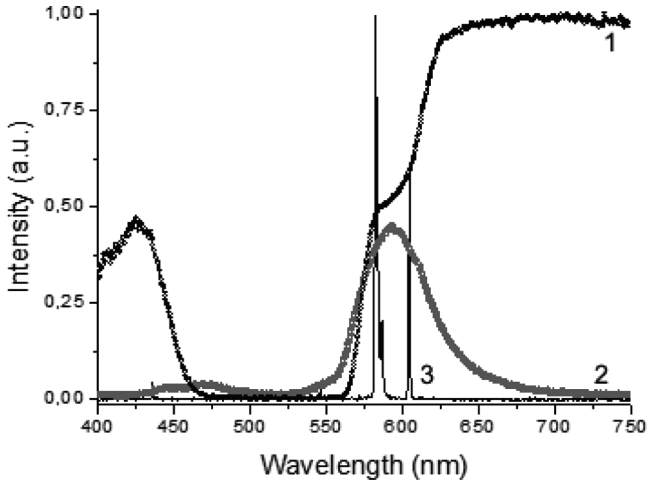

FIGURE 2 Transmission spectrum of the cholesteric (1, black circles), fluorescence of the dye (2, gray triangles) and lasing spectrum from the sandwich cell (3, solid line).

0.75 μJ/pulse. A photograph showing the cell and the screen with the laser beam spots is presented in Figure 5. The distance between the cell and the screen is 30 cm. Also note that simple float glass plates

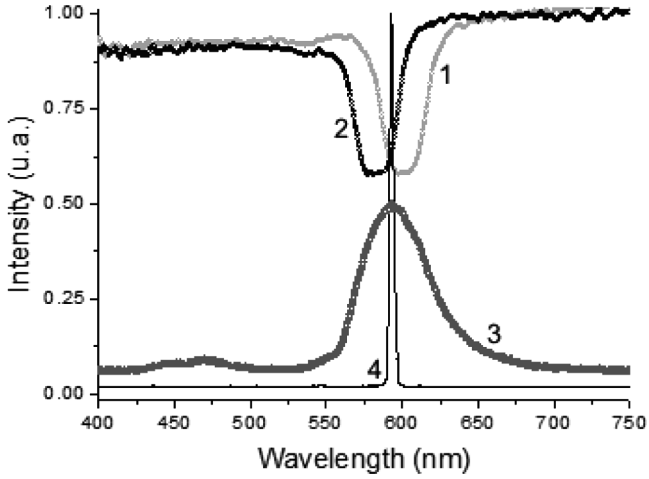

FIGURE 3 Transmission spectra of the two cholesterics with different pitches (1 and 2, black and light gray circles), fluorescence of the dye (3, gray triangles) and lasing spectrum from the sandwich cell (4, solid line).

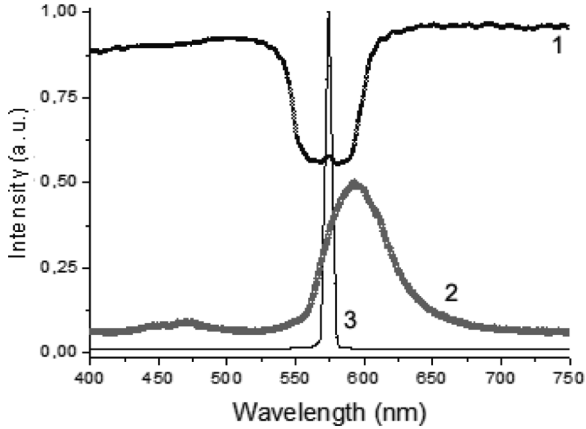

FIGURE 4 Sum of the transmission spectra of the two cholesterics (1, black circles), fluorescence of the dye (2, gray triangles) and lasing spectrum from the sandwich cell (3, solid line).

were used in our experiments. Obviously, the quality of their surfaces is far from being ideal for optical purposes, but, in spite of this, the divergence of the laser beam is considerably lower than the one usually observed in conventional DD CLC lasers. The diameter of the laser beam is less than 10 cm at 1 m far from the cell, while a conventional DD CLC laser prepared using the same glass plates shows a beam divergence of approximately 40 cm at the same distance.

Besides, Rhodamine-6G, another dye Stilbene-420, whose absorption and emission peaks are located in UV and violet ranges, was exploited as well. In this case, the CLC pitches were set to get lasing

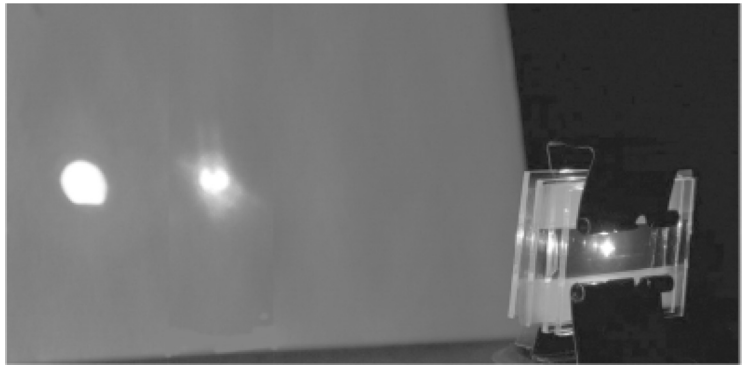

FIGURE 5 Laser emission from the sandwich cell.

in violet spectrum range and a nitrogen laser ($\lambda = 337$ nm) was used for pumping. Multimode lasing in the structures with equal CLC pitches and single mode one in the shifted pitch configuration were observed, confirming the general behavior.

CONCLUSION

We investigated defect mode lasing in three layer cells with separated CLC and dye solution layers. Multimode lasing was obtained when the helical pitches of the two CLC layers were the same. Single mode lasing was achieved shifting the CLCs pitches with respect to each other. Low divergence of the laser beam characterizes this kind of CLC lasers.

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