Wide band gap materials as a new tuning strategy for dye doped cholesteric liquid crystals laser

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Abstract: A new tuning strategy for mirror-less liquid crystals laser is presented. A three layer cell is prepared with two cholesteric layers sandwiching a layer containing an isotropic mixture of a photoluminescent dye. One of the chiral layers contains a wide band gap material while the second layer consists of a series of small band gap materials. Through the combination of these two layers, a set of mirrors that can selectively reflect different wavelengths is obtained. A different laser wavelength is emitted from different regions of the cell under the pumping beam irradiation.

OCIS codes: (230.3720) Liquid-crystal devices ; (160.3710) Liquid crystals.

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1. Introduction

Photonic band gap materials and devices which exhibit an ordered structure with periodic change of dielectric constant have attracted much attention from both theoretical and practical points of view. With a photonic band gap, photons can be strongly localized within the band gap or at the band edge, which paves the way for low-threshold lasers, micro-cavity lasers, optical diodes, and optical amplifiers [1]. With a self-organized chiral structure, cholesteric liquid crystal (CLC) is regarded as a one-dimensional (1D) photonic crystal.

In CLCs the period of helicoidal structure is equal to half the pitch p, and for light propagating along the helical axes, $p = \lambda_0 / n$, where λ_0 is the wavelength of the maximum reflection or the middle of selective reflection band (SRB) and n is the average refractive index $n = (n_e + n_o)/2$. The extraordinary and ordinary indices of refraction are denoted by n_e and n_o respectively. The full width at half maximum of the selective reflection band ($\Delta \lambda$) is equal to $p\Delta n$, where $\Delta n = n_e - n_o$ is the birefringence of a nematic layer perpendicular to the helix axis.

Cholesteric LCs possess several unique properties: periodic structure, 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, etc.). Finally, if the cholesteric LC contains a luminescent dopant, it becomes possible to build a tunable distributed feedback laser. Lasing from dye-doped CLC was obtained for the first time by Ilchishin et al. [2], and the consideration of the selective reflection band as a photonic band-gap [3] stimulated the investigation of lasing in several chiral materials. So far, low threshold laser action has been demonstrated not only in chiral nematic liquid crystals [4] but also in ferroelectric liquid crystals [5], TGB phases [6], cholesteric polymers [7], cholesteric elastomers [8]. Tunability of lasing in these systems was achieved varying the temperature [2], applying a mechanical stress [8] or an electric field [9], using photo-transformation effects of aromatic esters, azo and azoxy compounds [10-12] and by assembling cells with pitch gradient and with spatial distribution of different dyes [13,14].

Several attempts were made to optimize the lasing condition and the performance characteristics. A rotating cell was suggested to avoid deformation of the CLC texture and bleaching of the dye due to the high energy of the pumping beam [15]. An enhancement of lasing efficiency was recently observed using a CLC reflector in a dye doped CLC laser [16,17] or assembling a multilayer system that allows the separation of the cholesteric liquid crystal from the active medium [18]. The use of a CLC layer as a resonator – mirror was also proposed as an improvement for conventional lasers [19,20].

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Here we present two new strategies to finely tune the laser emission and to widen the range of the emitted wavelengths (420 nm - 790 nm) using a multilayer system.

The multilayer system consists in two separate cholesteric layers sandwiching a mixture containing a luminophore, which can also be not soluble in liquid crystals. The novelty of this system relies in the materials used for the cholesteric liquid crystal layers. One cholesteric layer contains a highly birefringent nematic material and shows a wide photonic band gap. For the other cholesteric layer two different solutions are investigated: a mixture containing a photosensitive chiral compound and a mixture containing a common chiral dopant.

The separation of the active medium from the cholesteric liquid crystal matrices allows: to avoid the degradation of the CLC structure caused by the absorption of the pumping energy and to use the optimal thickness both for the CLC layers and for the dye solution layer.

2. Experimental

For the experiments both commercial and specially prepared materials have been used. To obtain the wide photonic band gap layer, BL-006 or alternatively BL-090, with Δn approximately 0.3, as nematic compounds and MLC-6248 as optically active dopant have been used. To prepare the second cholesteric layer, MLC-6816 as nematic compound, ZLI-3786 as photosensitive chiral compound and RM-257 as photo- polymer (all the above materials supplied by Merck) and the Irgacure 2100 (Ciba) as photoiniziator have been used. As isotropic solvent glycerol has been used and as luminescent dyes ADS680HO (American Dye Source), not soluble in liquid crystals, and a specially prepared luminophore have been used.

The sandwich cell has been assembled as shown in Fig. 1, and it is composed by a first cholesteric layer containing a material with a well defined pitch and a second cholesteric layer containing chiral materials with different pitches.

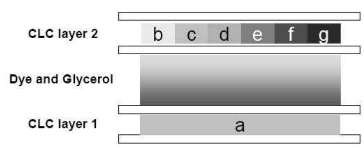

Fig. 1. Sketch of the sandwich cell.

The mixture containing dye and glycerol is sealed in a 1 mm central glass cell while the two cholesteric mixtures are sandwiched between two glass and quartz plates coated with rubbed PVA (polyvinyl alcohol) to obtain a homogeneous alignment of the CLC layers. The thickness of the two cholesteric layers is 5μm.

In order to tune the laser emission, two different strategies are exploited, the first one is based on the photo-transformation properties of the chiral compound ZLI-3786. The chemical structure of this material is identical to the one of ZLI-811 (Merck) except that they are optical antipodes [21], i.e. they possess a different handedness. ZLI-3786, like ZLI-811 [10,13], undergoes a photo-transformation if irradiated at wavelengths shorter than 300 nm. This transformation is a photo-Fries rearrangement, which is a well known phenomenon for aromatic esters. Its occurrence is revealed by the effect produced on the cholesteric helical pitch, which changes with the exposure time.

To create a pitch gradient within the CLC layer containing ZLI-3786, distinct regions of the layer have been exposed to UV for different times using a customized mask. The addition to the mixture of a photopolymer and a photoiniziator is needed to stabilize the cholesteric structure. The used illumination times have been from 1 up to 5 minutes, to have the helical pitch varying continuously within the second cholesteric layer.

Figure 2(b) shows the transmission spectrum for the mixture contained in the second cholesteric layer 99%[78% MLC-6816 + 22% ZLI-3786] + 1%[99% RM-257 + 1% Irgacure

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2100]. On illuminating the second cell with a 100W mercury lamp, a process that results in a stable shift of the selective reflection peak is induced. Figure 2 shows the transmission spectra of the mixture when it is irradiated for 1 min (c), 2 min (d), 3 min (e), 4 min (f) and 5 min (g) respectively. The band gap shifts towards longer wavelengths with increasing exposure time, more than 100nm, indicating an elongation of the helix.

As first cholesteric layer the following mixture has been used: 78% BL-006 + 22% MLC-6248. The transmission spectrum of the wide band gap material is shown in Fig. 2(a), the gap width is about 150nm. For a perfect cholesteric texture the reflection is 50%, in the present case for the wide band gap layer is around 45% and for the narrow band gap cholesterics it is around 40% .

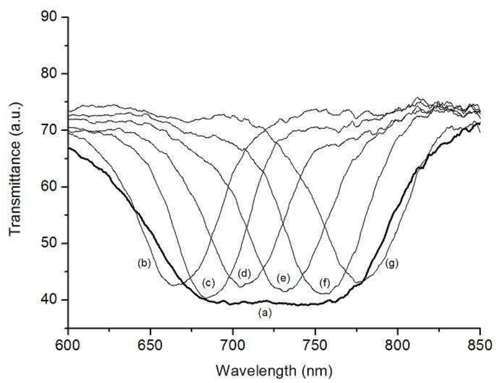

Fig. 2

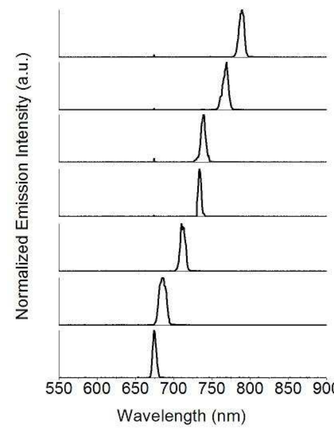

Fig. 3

Fig. 2 Transmission spectra of the wide band gap cholesteric (a) 78% BL-006 + 22% MLC-6248 and of the small band gap cholesterics obtained illuminating with a UV lamp different regions of a 99% [78% MLC-6816 + 22% ZLI-3786] + 1% [99% RM-257 +1% Irgacure 2100] mixture for 0 min (b), 1 minute (c), 2 min (d), 3 min (e), 4 min (f) and 5 min (g).

Fig. 3. Laser emission in the near infrared range.

Therefore we have a series of very narrow reflective band gaps from one side of the cell containing the luminescent dye and a wide photonic band-gap from the opposite side (Fig. 2).

Depending on the position of the pumping beam on the cell, one of the small band gap materials is selected. The dye emitted photons, whose wavelength is in the narrow range where the wide band gap and the selected smaller one overlap, undergo in optical resonance giving rise to a lasing effect.

In order to test the laser tuning, the cell has been placed on a translation stage and the sample has been moved in the plane orthogonal to the pumping laser beam. A nitrogen laser has been used as exciting source (VSL-337ND-S, Spectra Physics). The emitted laser beam from different regions of the sandwich cell is then investigated, the different laser wavelengths are shown in Fig. 3. Using a dye not soluble in liquid crystals, a laser emission in the near infrared at 790 nm has been observed for the first time.

The average laser width for every emitted wavelength is around 9 nm (Figs. 2,3) and it is related to the width of the narrow band gap cholesterics. The measurements of the laser lines width is limited by the spectral resolution of the experimental setup: an optical fiber coupled to a spectrometer (AVS-S2000, Avantes) having resolution ~1.4 nm. Moreover the characteristic properties of lasing depend on the quality of the CLC texture, on the change of this texture under the influence of the exciting laser beam and on the cells assembly to obtain the pitch gradient.

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The second strategy to obtain the modulation of the photonic band gap position of the second chiral layer relies on the use of a series of CLC mixtures in which the chiral compound concentration changes. Three mixtures with different concentrations of the optically active dopant have been prepared: 90.5% MLC-6816 + 9.5% ZLI-4572, 89.5% MLC-6816 + 10.5% ZLI-4572, 89% MLC-6816 + 11% ZLI-4572. Their transmission spectra are shown in Fig. 4 (b), (c) and (d) respectively. The cell has been partially filled, by capillarity, with one of the three mixtures; filling has been then completed using in sequence the other two mixtures. After assembly, different wavelengths of the visible range are selectively reflected by the whole system.

In this cell as first cholesteric layer the following mixture has been used: 69.5% BL-090 + 31.5% MLC-6248. The transmission spectrum of the wide band gap material is shown in Fig. 4(a), the gap width is about 100 nm.

As previously described, the combination of the two cholesteric cells, creates a set of mirrors in the overlapped regions (Fig. 4).

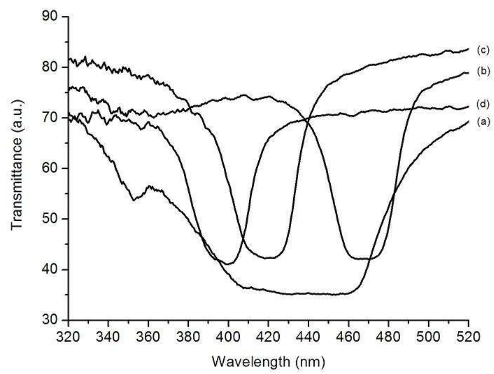

Fig. 4

Fig. 4. Transmission spectra of the wide band gap cholesteric (a) 69,5% BL-090 + 31,5% MLC-6248 and of the small band gap cholesterics obtained using different chiral dopant concentrations (b) 90.5% MLC-6816 + 9.5% ZLI-4572, (c) 89.5% MLC-6816 + 10.5% ZLI-4572, (d) 89% MLC-6816 + 11% ZLI-4572.

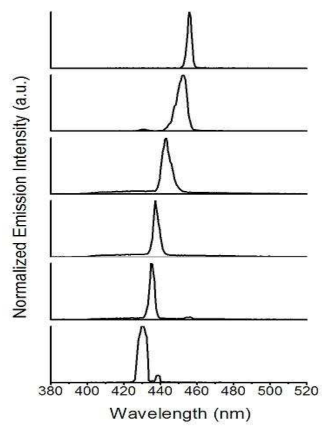

Fig. 5

Fig. 5. Laser emission in the ultraviolet range from 430 nm to 460 nm.

As luminescent material we used the specially prepared luminophore OF(mb). OF(mb) is an oligomer with chiral 2-(S)-methylbutyl pendant chains. This particular luminophore has been chosen as oligofluorenes (**OF**) have been reported to be very suitable blue-emitters for optical and electro-optical applications due to their good thermal and photo stability, and high emission efficiency. OF(mb) has been dissolved in glycerol.

Again the assembled cell has been shifted laterally with respect to the incoming pumping beam and the tuning of laser emission from 430 to 460 nm has been observed (Fig. 5).

In Fig. 5 (below) it is shown the presence of multimode emission. Usually lasing in three layered structures could be considered as a defect mode type and multimode lasing within the stop band with several emission peaks is typical for this case [22,23].

To obtain single mode lasing, additional conditions are needed: for three layer cells as described in [18], single mode lasing was achieved using cholesterics with different pitches, obtaining laser emission at the wavelength in which the two cholesteric selective reflection bands overlapped.

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3. Conclusions

We present a novel design to obtain widely tunable mirror-less lasers, that combines the reflective selection properties of a broad band gap cholesteric and of a series of narrow band gap cholesterics both assembled in one system.

The narrow band gap cholesterics can be obtained inducing in the same cell a variation of the helical pitch. Two strategies to vary the pitch, and then the position of the band gap are presented. The first strategy is based on the phototransformation of a photosensitive optically active dopant, while the second one relies on the variation of the concentration of a non photosensitive chiral compound.

The two cholesteric layers, the one with the wide band gap material and the other one containing the narrow band gap materials, are placed at the opposite sides of a three layered cell sandwiching a mixture of isotropic solvent and a photoluminescent dye.

When the pumping beam hits the cell a narrow band gap cholesteric is selected and lasing is obtained in the region where the gaps overlap.

From this new device we have obtained for the first time a wider modulation of the laser emitted wavelength from 420 nm to 790 nm. With a proper dye, longer laser emission wavelengths could be achieved that would be optimal for applications in biomedicine and cosmetics.

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