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Lasing Stability Enhancement in Dye Doped Cholesteric Liquid Crystals

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Lasing from dye doped cholesteric liquid crystal mixture was experimentally observed several years ago but in despite of all the efforts that have been done so far mirror less liquid crystals lasers are not yet suitable for practical applications. The main issues that have still to be addressed concern the efficiency, the stability and the tunability of the laser emission.

This work has been aimed to obtain an improvement in lasing stability from dye doped chiral mixtures confined in glass cells. Stability problems are mainly related to the absorption of the powerful pumping beam from the mixture that leads to a deformation of the cholesteric texture and a bleaching of the dye. To solve these problems we have investigated different surface coatings and new system configurations.

Keywords Cholesteric liquid crystal; lasing; photonic band gap

Introduction

Cholesteric liquid crystals selforganize in an helical structure that selectively reflects light. The selective reflection band (photonic band gap) is centered at the wavelength $\lambda = np$, where p is the helical pitch of the chiral liquid crystal and $n = (n_e + n_o)/2$ is the average refractive index of the cholesteric planes. If the helical pitch is altered, the color of the sample and the position of the selective reflection band will change.

Owing to the unique properties: supermolecular helicoidal periodic structure, 100% selective reflection of circularly polarized light and hence the ability to change their selective reflection wavelength changing external or internal factors, the cholesteric can be considered as resonator in laser emission from doped luminescent

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molecules. Laser emission from dye doped cholesteric liquid crystals was first obtained by Ilchishin *et al*. [1].

The consideration of the selective reflection band as a photonic band-gap [2] stimulated observations and investigations of lasing in several chiral structures, such as thermotropic [3–6] and lyotropic [7] CLCs, blue phases [8], chiral smectic C liquid crystals [9], polymer networks [10], elastomers [11], and glass forming CLCs [12]. Tunability of lasing in these systems [13,14] has been achieved varying the temperature [15], applying a mechanical stress [11] or an electric field [16].

The use of CLCs as laser source for applications is still an open issue. The main drawbacks associated with these devices are related to the efficiency and stability of the laser emission. This last problem is connected with phenomena generated by a powerful pumping beam: deformation of the CLC texture and degradation of the photo-luminescent dye molecules.

In the last years we have investigated several routes to overcome this scenario and obtain a more stable laser emission. In particular we have studied the possibility to use different materials as aligning substrates to have an optimal accommodation of the cholesteric helix, to move the cell assembly with respect to the pumping beam to prevent the beam from being absorbed from the same area of the cell for a too prolonged period of time, and to use a new cell assembly to separate the photoluminescent dye from the cholesteric materials.

Experiments and Results

In our investigations, an important matter has been the selection of proper chiral liquid crystals which present a good spectral overlapping of their photonic band edge with the high efficiency region of the dye fluorescence spectra. For our investigations MLC-6815/6816 as nematic liquid crystals and MLC-6247/6248 as optically active dopants have been used. These nematic materials as well as the chiral dopants are UV transparent and have a good thermal stability, the chiral compounds have the correct helical twisting power to obtain a pitch of the cholesteric helix comparable with the visible light wavelength. As photoluminescent dopants both commercial (DCM, Exciton) and specially prepared materials have been used.

As previously stated, a good cholesteric texture is the starting point to achieve a stable laser emission and this can be obtained using polymer coated surfaces to induce a planar alignment of the liquid crystal at the surface. The role of the surface treatment on the laser emission have been experimentally investigated. Cells have been prepared using glass plates covered with a thin layer of rubbed polymers: PVA (polyvinylalcohol) (Sigma Aldrich) and PI (LQ1800 from Hitachi Chemical). PVA has been prepared using a concentration of 0.5% wt. in water, spin coated and then cured in the oven for one hour at 110°C. PI has been prepared using a concentration of 2% wt. in 1-methyl-2-pyrrolidinone, spin coated and cured 1 h at 180°C and then an additional hour at 250°C.

The thin films have been rubbed separately using different velvet cloths and the cells have been assembled in two different configurations. In the first configuration glasses are aligned with the rubbing directions parallel to each other while in the second one the directions are antiparallel. Cells have been prepared using 40 μm thick mylar spacers.

Investigations on photoexcitation have been performed using as a source a nitrogen laser, Model VSL-337ND-S (Spectra-Physics). The pulse wavelength,

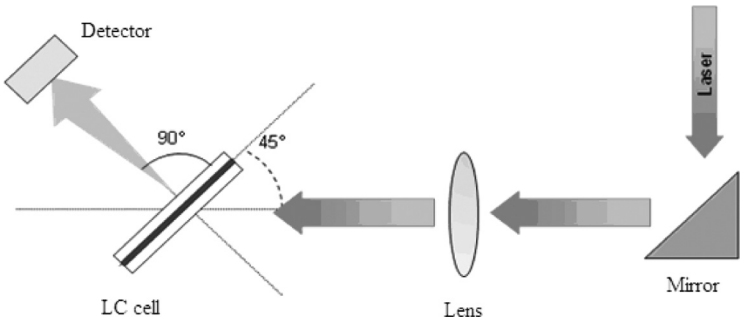

Figure 1. Experimental set-up for photo-excitation measurements.

width, and repetition rate are 337 nm, 4 ns, and 1–10 Hz respectively. The laser beam is focused by a lens ($f = 10$ cm) to reduce the spot size onto the cell to few hundreds of micrometers. The pumping beam hits the sample at 45° with respect to the cell normal, a usual experimental geometry for this kind of experiment (Fig. 1). An optical fiber coupled to the spectrometer Avantes Fiberoptics Model AVASPEC-2048, with 1.4 nm resolution, collects the light emitted from the sample.

Investigations have been performed filling cells with the mixture 99.6% (74.1%MLC-6816 + 25.9MLC-6248) + 0.4DCM. For all the measurements the power of the nitrogen laser has been set at about 1.5 μW, averaged on three seconds.

In Figure 2 it has been shown that the emission from the cell coated with antiparallel rubbed PI 2% is similarly intense with respect to the one from the cell coated

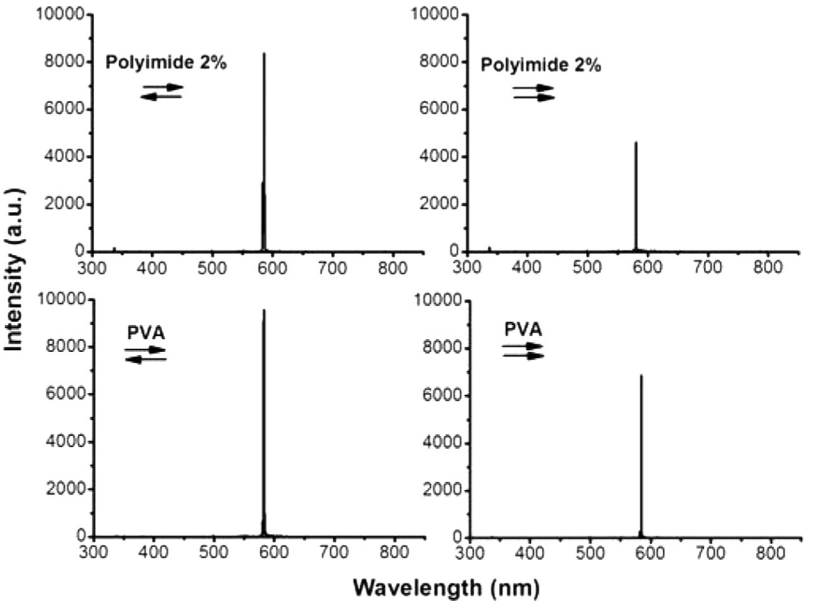

Figure 2. Laser emission from dye doped cholesteric liquid crystals mixture for cells with parallel and antiparallel rubbed PI 2% and PVA.

with antiparallel rubbed PVA. The emission from a cell coated with parallel rubbed PI 2% is about 30% less intense with respect to the one from the cell coated with PVA. The lasing emission from the cell containing antiparallel rubbed PVA shows to be the most efficient one. So far some polymers appear to be more suitable to obtain an homogeneous cholesteric texture and this is reflected in a more efficient emission from the DDCLC cell. Other polymers are currently under investigations to optimize the chiral structure. A theoretical interpretation is needed to relate the interaction polymer–liquid crystals at the interface to the configuration of the helix inside the cell and then to the intensity and stability of the laser emission.

Together with the damage of the cholesteric texture, the problem of dye molecules degradation is common to conventional dye lasers as well. To solve this problem, in these lasers the dye solvent is circulating continuously through the laser chamber avoiding the saturation effects from pumping [17]. For our cell assembly we cannot obtain a dye solvent circulation, but an important improvement in laser stability has been reached moving the cell or the impinging pumping beam one respect to the other. Figure 3 shows how the cell can be moved in the x-y plane and also can be rotated around the z axis. The main advantage for this configuration is that the exciting source does not hit the cell in the same area for a prolonged time.

In Figure 4 it is shown the average output power from a dye doped CLC (DDCLC) cell as a function of time for a cell that has been placed on a translational stage. The repetition rate of the pumping pulses was 7 Hz and the average pumping power was 45 μW. A power meter (Thermo Oriel Instruments) has been used to monitor laser emission. It must be noticed that an average power, averaged during several seconds of measurement (not per single pulse), has been measured.

The dye effectively absorbs the nitrogen laser pumping beam. Nevertheless, the measurements show that the lasing power of a moving cell remains stable for more than 10 minutes under the continuous irradiation of the pumping beam. Figure 4 shows the average output power for a DDCLC cell that moves in front of the

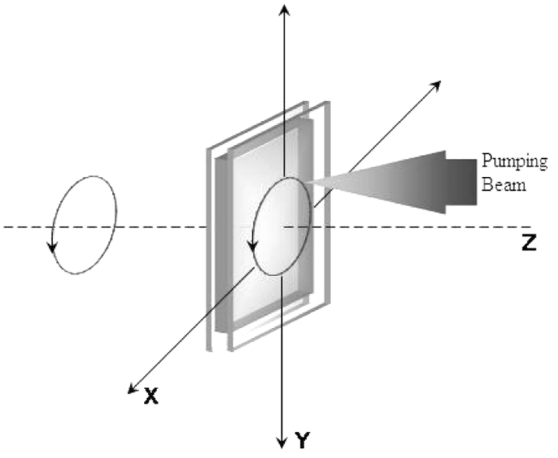

Figure 3. Sketch of the translational and rotational movements of the cell with respect to the pump source.

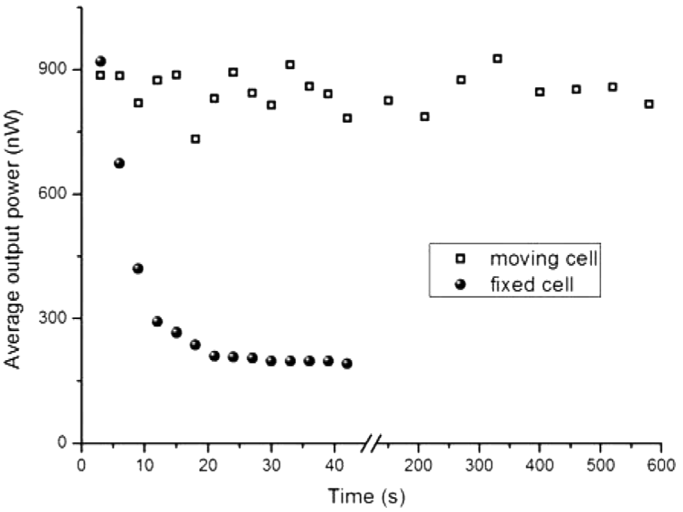

Figure 4. Average output power as a function of time from a DDCLC cell.

impinging beam and for a cell that is fixed. As it is shown the laser beam intensity decreases rapidly if the beam hits continuously the same spot on the cell. Other measurements, not reported here, show that the laser emission from a moving cell can be stable for hours [18].

Sufficient slackening of degradation of the planar CLC structure and the luminescent dye in the CLC lasers has been achieved rotating and/or translating the CLC cell. Again, as it is shown in Figure 4, fluctuations in laser emission are not completed avoided. The remaining lasing instability for a moving cell is mainly expressed by the laser fluctuations which are due to the spatial homogeneity of the CLC structure.

The homogeneity of the cholesteric texture can be improved using the proper aligning substrates, as previously described, but also using a new cell configuration to separate the active photoluminescent compound from the cholesteric matrices [19]. In the new device a layer containing a mixture of an isotropic solvent and the photoluminescent dye has been placed in the central part of a sandwich cell, while the outer cells contain chiral mixtures. These two last mixtures have been prepared different from each other to achieve single-mode lasing, and their transmission spectra are shown in Figure 5, curves a and b, their pitches have been shifted in such a way that only the edges overlap. The active medium is prepared using a solution of dye dissolved in glycerol, its luminescence spectrum is also shown in Figure 5, curve c. In this experiment the CLC pitches were chosen to set the lasing peak near the maximum of the dye luminescence peak as shown in Figure 5, curve d.

This peculiar cell assembly, at the same time, allows to have a minor damage of the cholesteric textures and to eventually use dyes which are not soluble in liquid crystals. Further, this geometry allows to optimise the thickness for the different layers. As an example for the active layer the cell thickness was several hundreds of microns while each cholesteric layer was a tens of microns thick. The main drawback of this system is the not easy assembly of the sample, nevertheless this system is very promising for further developments.

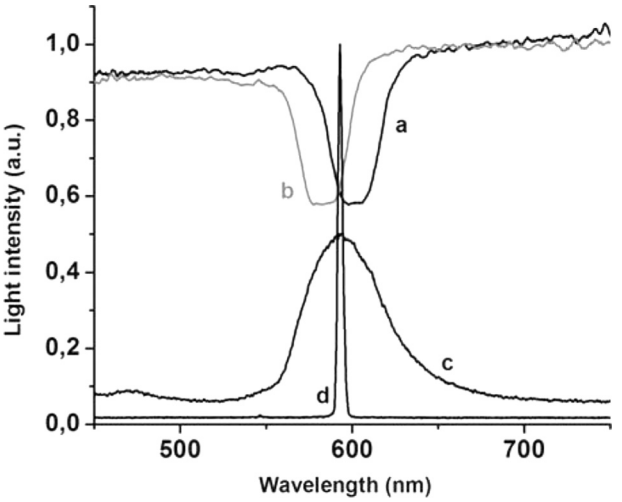

Figure 5. Transmission spectra (a, b) of the cholesteric mixtures, luminescence spectrum of the dye-glycerol mixture (c) and lasing peak (d).

Conclusions

We investigated different solutions to improve stability in lasing emission from DDCLC mixtures. We found that suitable surface treatments, cell assembly and a translational/rotational movement of the cell with respect to the pumping beam can help in obtaining a quite stable intensity of laser emission in time. These results have been used to prepare a prototype of a laser system, able to emit several

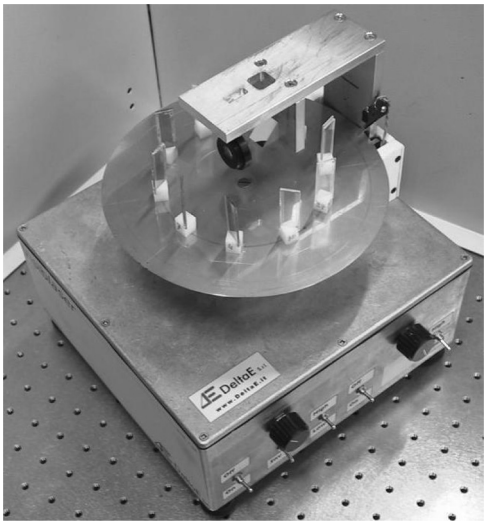

Figure 6. Laser prototype emitting at different wavelengths. A rotating plate allows to select a DDCLC cell which emits the appropriate light wavelength. The plate is also equipped with a translational movement that moves the selected cell in front of the pumping beam. The pump source is a nitrogen laser.

wavelengths in the visible range (Fig. 6). This prototype is a new light source suitable for light scattering experiments in biological systems [20].

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