Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl20

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Version of record first published: 22 Sep 2010

To cite this article: Hiroyuki Yoshida, Chee Heng Lee, Akihiko Fujii & Masanori Ozaki (2007): Tunable Chiral Photonic Defect Modes in Locally Polymerized Cholesteric Liquid Crystals, Molecular Crystals and Liquid Crystals, 477:1, 255/[749]-262/[756]

To link to this article: http://dx.doi.org/10.1080/15421400701688260

Mol. Cryst. Liq. Cryst., Vol. 477, pp. 255/[749]–262/[756], 2007

ISSN: 1542-1406 print/1563-5287 online
DOI: 10.1080/15421400701688260

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A structural defect is introduced in the helical periodic lattice of a cholesteric liquid crystal, a well-known pseudo one-dimensional photonic band-gap material. The method utilizes an ultra-fast pulse laser to induce two-photon polymerization in a photo-polymerizable cholesteric liquid crystal material to directly fabricate regions with different pitch lengths. Single or multiple defect modes were exhibited in the selective reflection band of the cholesteric liquid crystal, depending on the fabrication conditions of the defect structure. Optically pumped laser-action experiments showed that lasing from the single defect-mode had the smallest threshold compared to other lasing modes.

Keywords: cholesteric liquid crystal; defect mode; lasing; photonic band-gap

Cholesteric Liquid Crystals (ChLCs) are a kind of liquid crystals in which their so-called director, defining the averaged molecular orientation, possess a helical periodicity along a particular helical axis with a periodicity called the pitch, p. They thus form a twisted anisotropic medium (with ordinary and extraordinary refractive indices n_o and n_e) with their dielectric tensor varying periodically along the helical axis, resulting in a selective reflection band for light with the same circular polarization as the material itself, over a wavelength range given by

We thank Meck KGaA for providing the photo-polymeric cholesteric liquid crystal materials. This work is partly supported by a Grant-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology of Japan.

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$\lambda \sim n_o p - \underline{n}_e p$ [1]. Much attention has recently focused on the photonic properties of such flexible, self-organizing photonic band-gap (PBG) materials [2–4]. They have been utilized as tunable distributed feedback dye-lasers [5,6], and the effect of introducing structural defects in the helical lattice has been studied extensively, both from theoretical and experimental views [7–14].

A characteristic defect that has been introduced thus far in ChLCs is the twist defect mode (TDM) [12], which is a discontinuous shift introduced in the director distribution of the ChLC. This was realized experimentally by stacking two photo-polymerized ChLC (PChLC) films so that the liquid crystal director experiences a jump at the boundary of the two PChLC films [13,14], and a transmission peak exhibiting laser action at a lowered threshold than at the reflection band-edge was observed due to light-localization at the twist defect site. The technique to stack PChLCs is simple to implement into other structures, for example, combining them with non-polymeric materials: to date, structures which show fascinating properties like the optical diode effect [7] or low-threshold lasing [8,9] have been reported. However, the stacking technique is a top-down method which limits the size of the structure to be fabricated, and only structures with defect layers of few to several μms have been reported thus far in combined PChLC structures.

In this study, we employ an experimental method to introduce a structural defect in the helical structure of ChLCs by a bottom-up method. The principle of the fabrication method is shown in Figure 1. The ultra-fast pulse laser is tightly focused in the photo-polymerizable ChLC material, near the substrate surface within the LC cell.

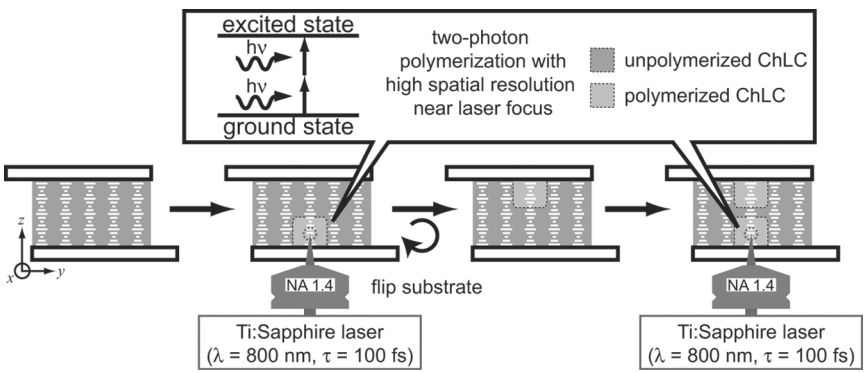

FIGURE 1 A schematic illustration of the introduction process of structural defects in cholesteric liquid crystals.

Two-photon polymerization occurs in the vicinity of the laser focus where the photon density is high, and a PChLC region is formed. The sample is then flipped over and the laser-lithography process is performed again on the opposite surface of the liquid crystal cell, resulting in a hybrid structure where an unpolymerized ChLC region is left between two PChLC films at the cell substrates. The unpolymerized ChLC region should therefore act as a structural defect introduced in the PChLC lattice. Since the dimension of the PChLC film is limited only by the spatial resolution of two-photon polymerization which may go down to 100 nm [15], high-resolution manipulation of the polymerizing region of ChLCs should be achieved by this method.

In experiment, 100-fs light with $\lambda = 800\,\text{nm}$ and repetition rate of 80 MHz from a Ti:Sapphire laser (Spectra Physics: Maitai) was focused through an objective lens with N.A. 1.4 on a 1 wt% DCM dye-doped (Exciton) PChLC material (Merck KGaA: 56 wt% 02-595 + 44 wt% 02-596) infiltrated in a cell of approximately 6-μm thickness. A rectangular area of size $146.2\,\mu\text{m} \times 146.2\,\mu\text{m}$ was scanned with a scan-line resolution of 2048 scan-lines per scan-area, at a scan-speed of $126\,\mu\text{s}\,\mu\text{m}^{-2}$. Figure 2 shows the thickness of the fabricated PChLC films as a function of the power density of the laser. The PChLC film thickness increases linearly upon increasing the power density: our experimental configuration allowed a vertical polymerization resolution of a few hundred nanometers to be attained.

Figure 3 describes the modification of helix caused by the laser-lithography process. The rectangular area shown in the transmission

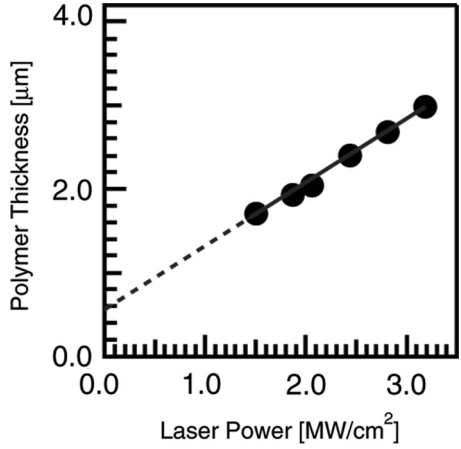

FIGURE 2 The thickness of the PChLC films fabricated by laser-lithography as a function of the power density of the Ti:Sapphire laser.

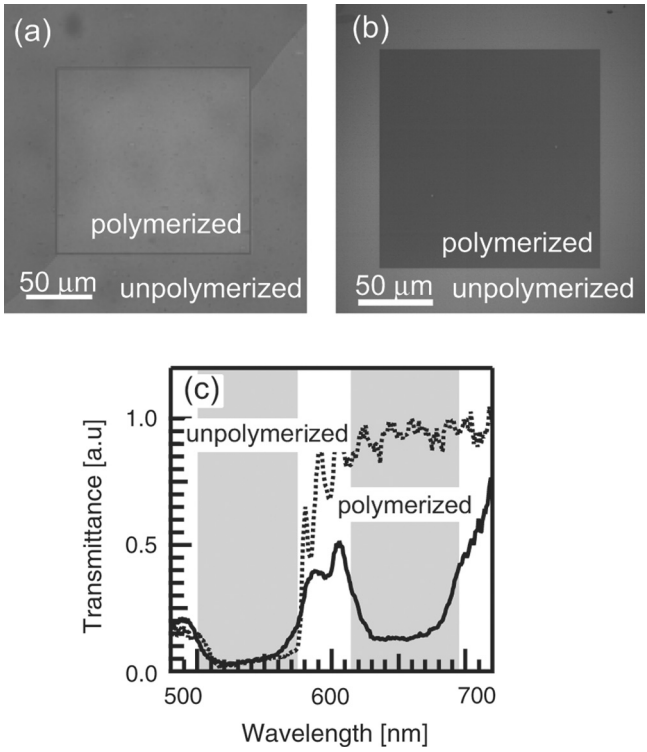

FIGURE 3 (a) A transmission micrograph upon RCP light incidence and (b) confocal laser micrograph of the PChLC material after laser-lithography had been performed the rectangular region indicates the polymerized region, i.e. the area scanned by the laser. (c) The transmission spectra of the unpolymerized ChLC and the polymerized ChLC upon RCP light incidence.

micrograph (Fig. 3(a)) or the confocal laser micrograph (Fig. 3(b)) is the area in which laser-lithography had been performed and a polymerized ChLC region is formed. A distinct color change, which occurs gradually over a time period of several minutes to tens of minutes after laser-lithography, is observed at the polymerized region. A comparison of the transmission spectra of the two regions (for a sample with laser-lithography performed at $1.58\,\mathrm{MW\,cm^{-2}}$, Fig. 3 (c)) shows that the selective reflection band shifted to the longer wavelength region, from $\lambda_c \sim p(n_e + n_o)/2 \sim 533\,\mathrm{nm}$ at the unpolymerized region to $\lambda_c \sim 654\,\mathrm{nm}$ at the polymerized region, corresponding to a pitch elongation of about 20%. We believe this anomalous helix elongation is caused by the diffusion of the chiral dopant out of the polymer matrix: the confocal laser scanning micrograph (Fig. 3(b)) shows a

decrease of fluorescence upon polymerization of the ChLC attributed to the diffusion of the DCM dye out from the polymer matrix, and the chiral dopant may also have come out of the polymer matrix by a similar phenomenon. Because of the helix elongation of the polymerized ChLC, the system becomes a hybrid structure with two PChLC films with an elongated pitch sandwiching the unpolymerized ChLC with a shorter pitch.

Figure 4 shows the transmittance spectra for right circularly polarized (RCP) light of the fabricated ChLC defect structures, with different laser intensities for the fabrication of the PChLC films: 3.0, 2.6 and 1.6 $MW\,cm^{-2}$. In each structure, defect modes with a transmittance peak are observed within the selective reflection band of the ChLC. For each power density configuration, the approximate PChLC film thicknesses are 2.8 μm, 2.4 μm and 1.75 μm, which yield a defect region with approximate widths 1.4 μm, 2.2 μm and 3.5 μm. Since the number of the defect modes permitted within the selective reflection band is determined by the thickness of the defect layer, which is the unpolymerized ChLC region, different numbers of defect modes are realized for each structure. This means that the defect modes can be manipulated by configuring the laser intensity to fabricate PChLC films with different thickness.

The lasing experiment from the fabricated structures was performed with second harmonic pulses ($\lambda = 532$ nm, 10 ns, 10 Hz) from a Q-switched Nd: YAG laser (Spectra Physics: Quanta-Ray INDI) as the pump source. The laser was focused on a spot approximately 10 μm in diameter, and the emission spectra were measured with a multichannel spectrometer (Hamamatsu Photonics: PMA-11) with a resolution limit of 2 nm. Figure 5 shows the emission spectra at high and low pumping energies of the non-deformed ChLC and the ChLC structures with a defect to exhibit either a single or multiple defect-modes, along with the corresponding transmittance spectra. In the non-deformed ChLC, laser emission is observed at 590 nm, where the longer selective reflection band-edge is positioned. On the other hand, for the ChLC defect structures, laser emission is exhibited from the defect-mode wavelengths, which is at 628 nm for the single-defect mode structure and 623 nm for the multi-defect mode structure. The lasing thresholds for the three lasing modes are approximately 31.4 mJ per pulse (33.8 $mJ\,cm^{-2}$ per pulse), 15.6 mJ per pulse (16.7 $mJ\,cm^{-2}$ per pulse) and 18.8 mJ per pulse (20.2 $mJ\,cm^{-2}$ per pulse) for the non-deformed ChLC, the single-defect and multi-defect mode structures respectively. Considering the fluorescence peak of the DCM dye at 600 nm being close to the selective reflection band-edge wavelength, the almost two-fold improvement in the lasing

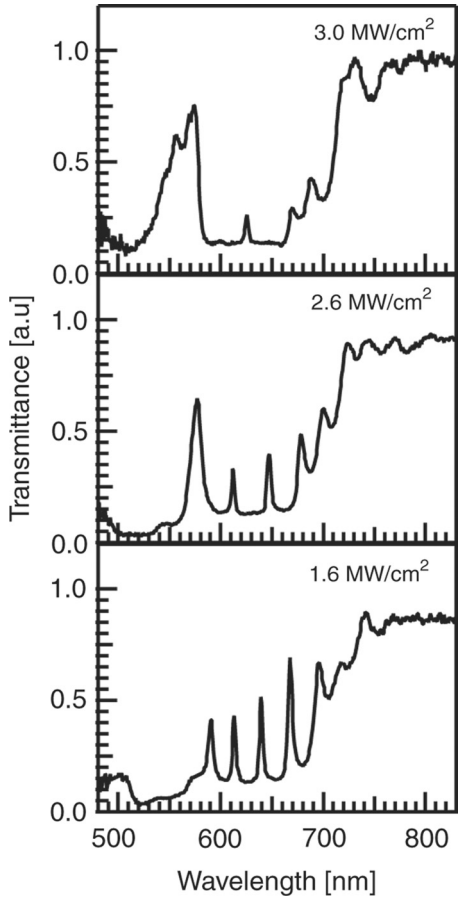

FIGURE 4 The transmittance spectra of the fabricated structures where the unpolymerized region placed between the polymerized PChLC films fabricated at different laser power densities act as structural defects. The difference in the defect widths leads to different number of defect modes being exhibited in the selective reflection band.

threshold from the defect modes is a striking result, indicating the high-Q of the defect structure functioning as a cavity. The lower lasing threshold from the single-defect mode structure compared to the multi-defect mode structure may be attributed to a more efficient lasing from the single-defect mode where the photon density of states is confined in one wavelength, whereas in the multi-defect mode structure the photon density of states is distributed over the multiple defect mode wavelengths.

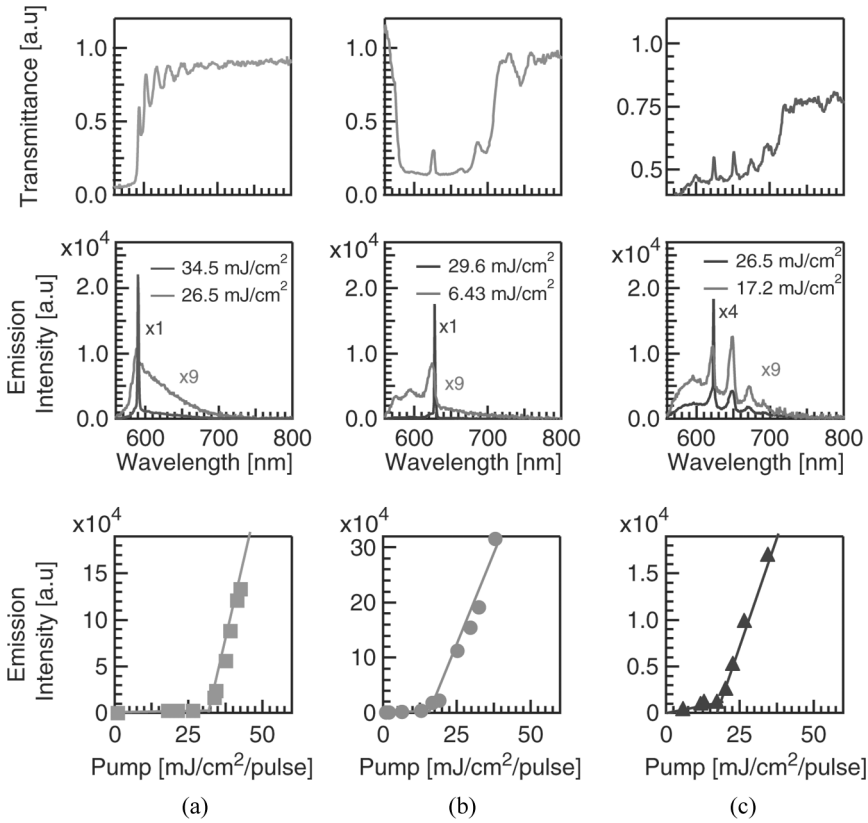

FIGURE 5 The emission spectra from the doped DCM dye in the non-deformed ChLC and the structure with the defect-modes at high and low pump energies, their threshold characteristics and their corresponding transmittance spectra (Figs. (a) and (b) are measurements upon RCP incidence while (c) is for linearly polarized light incidence).

In conclusion, we employed a bottom-up method to introduce defects of arbitrary size in ChLCs and proved its effectiveness by controlling the number of defect modes permitted in the selective reflection band of ChLCs. We also realized laser action with a lowered threshold from the defect-mode wavelengths, due to the cavity effect caused by the defect structures. We believe utilization of the photonic propertied induced by nano-scale manipulation of self-organizing structures lead to realization of intelligent devices useful for photonic circuits and communication systems.

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