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Tunable single photonic defect-mode in cholesteric liquid crystals with laser-induced local modifications of helix

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The authors demonstrate a tunable single photonic defect-mode in a single cholesteric liquid crystal material based on a structural defect introduced by local modification of the helix. An unpolymerized region of cholesteric liquid crystal acting as the defect was left between two polymerized regions via a two-photon excitation laser-lithography process. Upon polymerization, the cholesteric liquid crystal helix elongated and became thermally stable, and a single photonic defect mode was exhibited due to the contrast in the helix pitch at the defect. The defect mode showed tunability upon heating, and a 36 nm redshift was seen over a temperature range of 30 °C. © 2006 American Institute of Physics. [DOI: 10.1063/1.2400070]

Cholesteric liquid crystals (ChLCs) with chiral constituents spontaneously form a helical periodic structure with a periodicity called the pitch. Because of the periodic structure, they exhibit selective reflection (SR), a photonic band gap for circularly polarized light with the same handedness as the material itself.¹ The photonic properties of these self-organizing, pseudo-one-dimensional photonic band-gap materials have been investigated extensively. Low-threshold laser action has been reported from the SR band edge of ChLCs,² and tuning of the lasing wavelength has been reported, utilizing their ability to change the pitch by external fields such as heat³ or optical fields.⁴ On the other hand, laser action with high thermal stability has been reported in photopolymerizable ChLCs (PChLCs) in which the helical structure has been stabilized by polymerization.⁵

Introduction of structural defects in ChLCs has also attracted notable interest. Numerous defect structures had been proposed by stacking PChLC films, due to its simple fabrication method. Low-threshold lasing from the twist defect mode was demonstrated in a stacked configuration of two PChLC films,^{6,7} while hetero-ChLC structures with a nematic LC (Refs. 8 and 9) or a ChLC (Ref. 10) defect layer placed between two PChLC films were reported to show interesting optical properties such as the optical diode effect. In this study, we present a nonstacking process to introduce structural defects in a single PChLC material, and demonstrate a tunable single photonic defect mode, which has not yet been demonstrated by the stacking method.

Figure 1 shows the experimental procedure of introducing the structural defect. The sample cell is prepared by aligning a PChLC material with the helical axis perpendicular to the substrate. We irradiate 100 fs pulses of a Ti:sapphire laser (Spectra Physics: Maitai) at $\lambda = 800$ nm and repetition rate of 80 MHz on the sample positioned on a confocal laser scanning microscope (CLSM) system (Zeiss: LSM-510). The laser is focused by a high-numerical aperture (NA) objective lens (oil immersion, 63 \times , NA of 1.4), and is controlled by a galvanometer to scan a rectangular area of approximately 146.2 \times 146.2 μm^2 . In the vicinity of the laser

focus where the photon density is high, a two-photon absorption process is induced, polymerizing a finite region within the sample cell [Fig. 1(b)]. The unpolymerized region left between two photopolymerized ChLC regions formed at both substrates acts as the structural defect [Fig. 1(e)].

The sample used in experiment was prepared by mixing two right-handed PChLC mixtures, 02-595 and 02-596 (Merck) at a ratio of 56:44 (which polymerizes at $\lambda \sim 400$ nm), after which 1 wt % of 4-(dicyanomethylene)-2-methyl-6-(4-dimethylaminostyryl)-4H-pyran (DCM) (Exci-

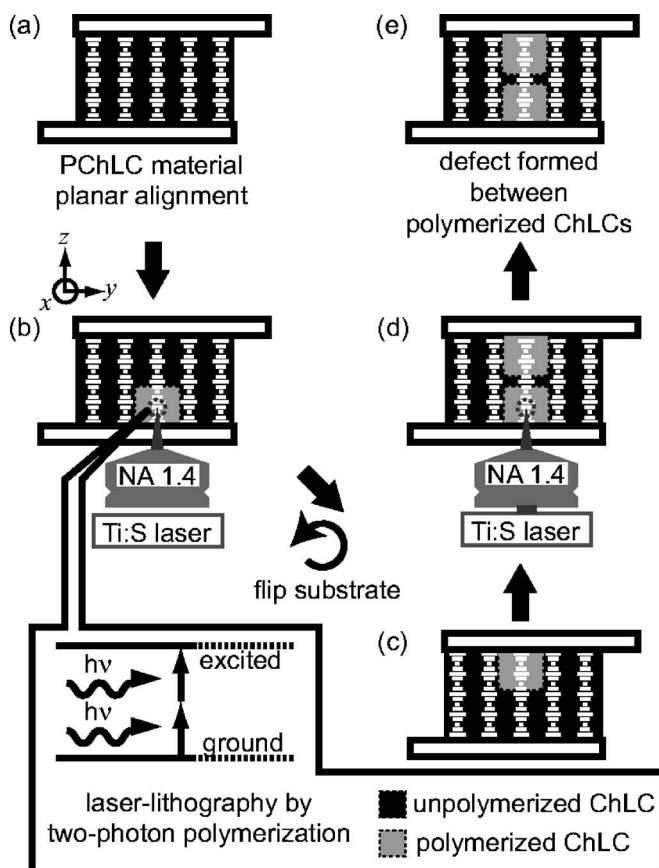


FIG. 1. Schematic illustration of the experimental procedure to introduce a structural defect in ChLCs.

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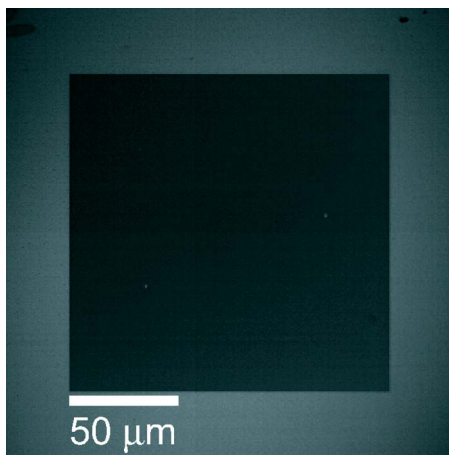


FIG. 2. (Color online) CLSM image of the sample after a laser scan was performed in the square area shown in image. The fluorescence was seen to diminish upon polymerization.

ton) dye was dissolved for observation purposes. The SR band edge is at $\lambda \sim 600$ nm, so it can be distinguished from the absorption of the DCM dye. The sample was infiltrated in a cell with cell gap of approximately $5.7 \mu\text{m}$ at a temperature slightly above the clearing point ($\sim 106.5^\circ\text{C}$) and slowly cooled to 45°C where it is in the ChLC phase (laser lithography was also performed at 45°C because the sample crystallizes at room temperature). Laser lithography was performed at a laser intensity of 0.61 GW cm^{-2} and scan speed of $126 \mu\text{m } \mu\text{s}^{-2}$. Only a single scan was required for polymerization: a CLSM image taken after the scan (Fig. 2, taken at 0.1 GW cm^{-2}) showed diminished fluorescence of the DCM dye in the polymerized region and could be distinguished from the unpolymerized ChLC. The polymer thickness was approximately $2.5 \mu\text{m}$ yielding an $\sim 0.7 \mu\text{m}$ wide unpolymerized region acting as the defect.

We first characterized the changes induced in the PChLC upon inducing local photopolymerization. One could expect a slight change in the refractive index of the material after polymerization, but to our surprise an anomalous elongation of the helix caused the optical characteristics to change drastically. Figure 3 shows the transmittance spectra of the PChLC before and after single-side laser lithography [corresponding to Figs. 1(a) and 1(c) respectively], for right-circularly and left-circularly polarized lights, measured on a polarization microscope (Nikon: Eclipse E600 POL) with an objective with NA of 0.4 by a charge-coupled device spectrometer (Hamamatsu Photonics: PMA-11). When a finite ChLC region was polymerized, an extra SR band (shaded in graph) appeared at a longer wavelength region than for the unpolymerized ChLC. Numerical calculations based on the 4×4 matrix formulation¹¹ revealed that the two SR bands corresponded to pitch lengths of 331 and 403 nm (using $n_e = 1.72$ and $n_o = 1.53$), indicating that pitch elongation had occurred. We believe this anomalous elongation of helix is caused by the diffusion of the chiral dopant in the PChLC mixture from the polymerized region into the unpolymerized region for the following reasons: (i) the elongation of the helix occurred only when polymerization was induced locally by laser lithography (not when the whole material was polymerized uniformly by UV light), indicating that a finite polymerized region needs to be surrounded by an unpolymerized region for pitch elongation to occur, (ii) the dimin-

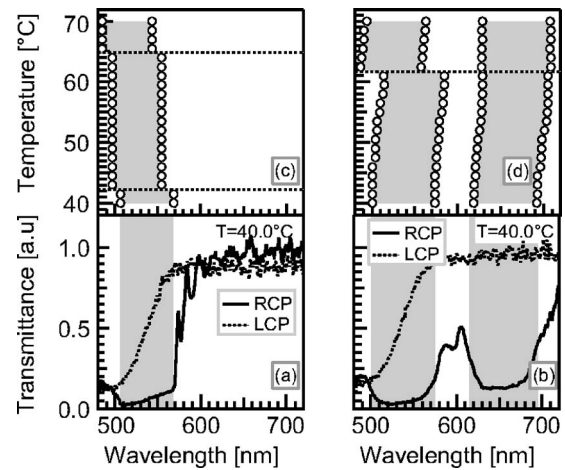


FIG. 3. Transmittance spectra at 40°C and the temperature dependence of the SR bands (shaded region) of the PChLC material [(a) and (c)] before laser lithography [Fig. 1(a)] and [(b) and (d)] after a polymerized region was formed on a single substrate [Fig. 1(c)]. The absorption of the DCM dye is observed at <550 nm.

ished DCM fluorescence from the polymerized region indicates that certain nonpolymeric components such as the DCM dye and chiral dopant in the PChLC mixture may have come out of the polymer matrix after polymerization, and (iii) the polymer slowly changed color after laser lithography, indicating a gradual process. Because there is a contrast in the pitch length in the polymerized (bulk) and unpolymerized (defect) regions, we can expect a defect mode to appear in the SR band.¹²

The temperature dependence of the SR bands [Figs. 3(c) and 3(d)] indicates higher heat stability in the polymerized ChLC. In the unpolymerized ChLC, a stepwise blueshift of the SR band is observed by heating, corresponding to a discrete shortening of the pitch due to an increase in the number of half pitches existing in the liquid crystal cell.³ On the other hand, the SR band of the polymerized ChLC does not show a discrete pitch shortening by heating, indicating that the helix became stable against the external heat. The stabilization is a result of the dense cross-linking of the monomers upon polymerization, which prevents the molecules to move freely to either wind or unwind.⁵ A tunable photonic defect mode should utilize this difference in the thermal response of the two regions. The gradual redshift observed in the SR bands in Fig. 3(d) may be due to the expansion of the polymer matrix upon heating, and the gradual unwinding of the ChLC at the polymer-LC interface with weaker anchoring. This is still under investigation along with the actual LC director distribution at the newly formed interface, but is contrary to the dynamics in Fig. 3(c) where there is strong anchoring and no gradual shift of the SR band is observed.

Figure 4(a) shows the transmittance spectrum of the structure with polymerized ChLC regions at both substrate surfaces of the sample cell [Fig. 1(e)]. A single defect mode is exhibited at $\lambda = 645$ nm within the SR band of the elongated helix of the polymerized ChLC, exhibited between 605 and 695 nm. Although the distribution of the LC director in this structure with newly formed surfaces still needs characterization and should be discussed elsewhere, a theoretical calculation of the transmittance spectrum using parameters described above gives good agreement with the experimental results, supporting that a single photonic defect mode was

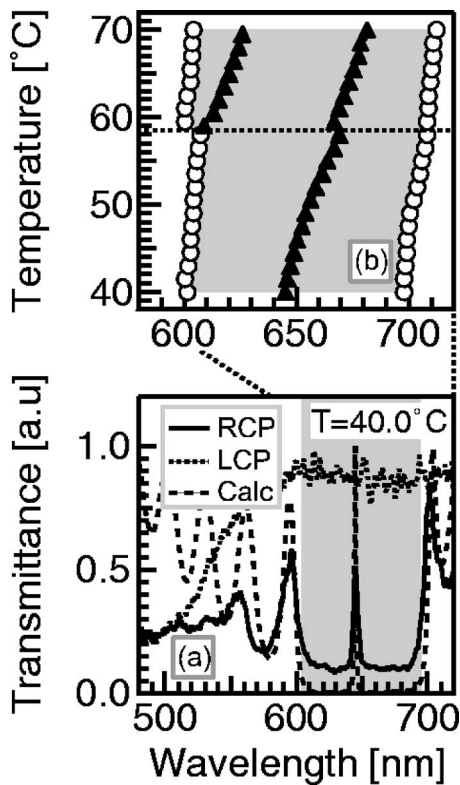


FIG. 4. (a) Transmittance spectra at 40 °C and (b) temperature dependence of the defect mode wavelength and the SR band (shaded gray) of the PChLC material after forming polymerized regions on both substrates [Fig. 1(e)].

realized from the unpolymerized ChLC acting as a defect introduced between two polymerized ChLC regions.

The tunability of the defect mode was demonstrated by heating the sample from 40 to 70 °C [Fig. 4(b)]. The defect mode wavelength redshifted upon heating, exhibiting a 36 nm shift over the 30 °C temperature range. The main cause of this shift may be the expansion of the polymer, diminishing the width of the defect. A different stepwise shift of the defect mode was observed at 59.0 °C, corresponding to a discrete shortening of the ChLC pitch at the defect in contrast to the bulk material. This may be caused by the ChLC at the defect favoring the pitch-shortened state at high temperature because of the surrounding ChLC with a discretely shortened pitch, although at the defect the unpolymerized ChLC has two polymer-LC interfaces with “weaker” anchoring. The increase in the contrast of the two

pitch lengths is also accountable for the second defect mode appearing in the SR band. Therefore, a tunable single photonic defect mode is achieved in a single PChLC material because of the laser-induced helix modulation.

In conclusion, we demonstrated a tunable single photonic defect mode in ChLCs based on inducing local photopolymerization by two-photon excitation laser lithography. The ChLC pitch elongated by a factor of about 20% and became thermally stable after laser lithography, thus creating a contrast in the pitch between the polymerized and the unpolymerized regions, enabling the structure act as a defect. We fabricated a structure with two polymerized layers sandwiching an unpolymerized region approximately $\sim 0.7 \mu\text{m}$ wide, in which a single defect mode was realized. The defect mode wavelength was variable by 36 nm upon heating the sample by 30 °C and causing an expansion of the polymer as well as a shortening of the ChLC pitch in the unpolymerized region. These optical devices may be used as tunable narrow band-pass filters or heat sensors in integrated optical circuits.

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