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Polarization characteristics of phase retardation defect mode lasing in polymeric cholesteric liquid crystals

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Abstract

We have studied the lasing characteristics of a dye-doped nematic layer sandwiched by two polymeric cholesteric liquid crystal (CLC) films as photonic band gap (PBG) materials. The nematic layer acts as a defect layer, the anisotropy of which brings about the following remarkable optical characteristics: (1) reflectance in the PBG region exceeds 50% due to the retardation effect, being unpredictable from a single CLC film; (2) efficient lasing occurs either at the defect mode wavelength or at the photonic band edge; and (3) the lasing emission due to both the defect mode and the photonic band edge mode contains both right- and left-circular polarizations, while the lasing emission from a dye-doped single CLC layer with a left-handed helix is left-circularly polarized.

Keywords: Photonic band gap; Polymer cholesteric liquid crystal; Defect mode; Lasing; Phase retardation; Polarization

1. Introduction

Crystals possessing a periodic modulation of their dielectric function, called photonic crystals (PCs), can inhibit certain frequencies of electromagnetic radiation from propagating through specific crystal orientations, i.e. photonic band gap (PBG) [1]. PCs are widely studied for their optical properties that allow manipulating the flux of light and making these materials good candidates for all optical signal processing. Particularly, cholesteric liquid crystals (CLCs) are one of the most interesting one-dimensional PC materials because of their nature of self-assembly. Light having the same handedness of circular polarization as that of the CLC helix cannot propagate through the CLCs in a frequency range corresponding to the PBG [2]. The same phenomenon has also been observed in certain kinds of insect cuticles [3]. Even more interesting is a beetle named *Plusiotis resplendens* that reflects both right- and left-circularly polarized light components [4]. This latter effect originates from a structure consisting of a unidirectional orientation of fibrils, that act as a λ/2 plate, sandwiched by two films with a left-handed helix (L-helix), that acts as a PBG structure. In this paper, we demonstrate lasing from photonic structures similar to those in the cuticle of *Plusiotis resplendens*.

The observation of spontaneous emission in dye-doped CLCs [5] has made lasing in CLCs become one of the most interesting areas. Lasing at the PBG edge has been reported in low molecular CLCs [6–8], polymeric cholesteric liquid crystals (PCLCs) [9], chiral smetic LCs [10] and the blue phase [11]. Among them, because of a reduced lasing threshold, higher quantum yield, improved emission [9] and good processibility, PCLC films are regarded as the best candidate of organic material for the formation of continuous wave lasing and organic diode lasers by charge injection, which are desirable in the view of practical applications.

Physical imperfections purposely introduced in periodic PC structures can cause an additional resonant mode

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(defect mode) inside PBG [1,12]. The use of defect modes has also been proposed for lasing at low threshold [12,13]. Generally, two kinds of configurations for generating defect modes in CLCs are suggested theoretically: introduction of an isotropic spacing layer in the middle of CLCs [14] and creation of a phase jump without any spacing in CLC structures [15–17]. Recently, Schmidtke et al. [18] experimentally demonstrated the defect mode lasing caused by a phase jump in CLCs and described its polarization characteristics. We recently demonstrated efficient lasing from an anisotropic defect layer in CLCs [19]. In this study, we investigated the polarization characteristics of the phase retardation defect mode emission, being different from that of single layer and twist defect mode systems in CLCs.

2. Experimental

For PCLCs, mixtures of two nematic liquid crystal (NLC) polymers (Nippon Oil Corporation) were used. One of the NLC polymers contains 25% chiral units in its chemical composition, giving right-handed helix (R-helix). By changing the mixing ratio of the two NLC polymers, the wavelength of PBG in PCLCs was controlled [20]. To fabricate PCLC films, a solution of the PCLC mixture was spin-coated on glass substrates with unidirectionally rubbed polyimide (AL 1254, JSR). After the coated PCLC films were cured for 2 min at 180 °C, we obtained well-aligned PCLC films. The cholesteric helical axis of the PCLC film was normal to the substrate surface and the thickness of the obtained PCLC film was about 1.8 μm. A pair of PCLC coated substrates were stacked and sealed to fabricate a vacant cell with a spacer, as shown in Fig. 1. A commercial monomeric mixture of NLCs (ZLI2293, Merck) was doped with a fluorescent polymeric dye [21] of 2 wt% and was introduced into cells fabricated by the PCLC films using a capillary action to form an anisotropic defect layer, as shown in Fig. 1. For comparison, we also prepared polymer-dye-doped simple CLC, consisting of a host NLC (Zli 2293, Merck) and 27.9 wt% of the chiral dopant (MLC 6247, Merck). The cell thickness was about 14 μm, and the L-helix [8] was formed with the helical axis along the substrate surfaces.

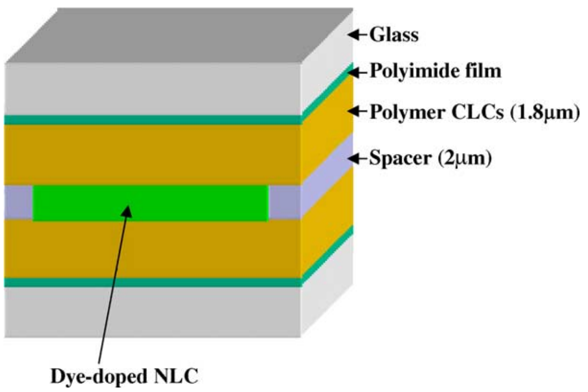

Fig. 1. Schematic structure of a CLC cell with a dye-doped NLC defect.

Reflectance spectra were observed using a microscope spectrometer (ORC, TFM-120AFT-PC). For the measurements of fluorescence and lasing spectra, a 440-nm pulsed laser beam from an optical parametric oscillator (Surelite OPO; HOYA Continuum) pumped by a third-harmonic light from a Nd:YAG laser was used for an optical pumping source. The pumping laser beam was focused on the sample surface at oblique incidence (about 30°) and the emission from the sample cell was collected by a lens and then detected by the multi-channel spectrometer (USB 2000:Oceon Optics, Inc.).

3. Results and discussions

Fig. 2 shows the measured reflectance spectrum from a fabricated sample cell (solid curve). The reflectance spectrum of a single PCLC film (dash-dot curve) is also shown for comparison. A striking phenomenon should be noted for the PCLCs with dye-doped NLC defect layer; i.e. the reflectance in the PBG region is higher than 50%, being quite different from other previously suggested defect modes in CLCs [19]. In contrast, the single PCLC film shows lower reflectance of about 40%, as expected.

A reflectance higher than 50% is caused by the phase retardation effect of the defect layer. Let us consider an ideal case like the cuticle of *Plusiotis resplendens* [4], that has a λ/2 plate between R-helix films, as mentioned above. Right-circular polarized (R-CP) light is reflected by the R-helix and left-circular polarized (L-CP) light passes through. The transmitted L-CP light is changed to R-CP light after passing through a λ/2 plate, then R-CP is again reflected by the second R-helix. Since this R-CP light is converted back to L-CP light by the λ/2 plate, it transmits through R-helix.

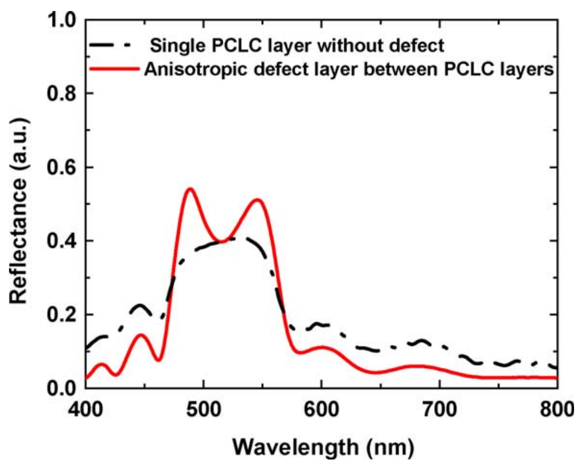

Fig. 2. The reflectance spectra of a single layer of CLC (dash-dot curve) and a CLC cell with a dye-doped NLC defect (solid curve).

Finally, 100% reflectance is obtained. In the present case, the thickness of the nematic layer is not controlled, so that the above effect is partially achieved, resulting in a reflectance higher than 50%, as shown in Fig. 2.

The other feature that should be noted is a notch of reflectance in the middle of the PBG region. This is a new type (retardation) of defect mode in the sense that the line shape is fairly broad in contrast to the normal defect mode which shows a sharp and deep dip [19]. The magnitude and shape of the reflectance can be controlled by adjusting the thickness and the anisotropy of NLC defect layers.

It is well known that polarized light of the same handedness as the cholesteric medium is suppressed inside the PBG and enhanced at the band edge of the PBG in single layer CLC film [22]. This is why lasing occurs at the edge of PBG and the lasing emission is circularly polarized with the same handedness as CLC. As for CLC films containing a 90° phase jump inside the helix, Kopp et al. [15] suggested theoretically that the polarization of light is affected by the film thickness. CP emission with the same handedness as the CLC helix is dominant below the crossover thickness, where equal contributions exist for L- and R-CP, and CP emission with opposite handedness as the CLC helix becomes dominant above the crossover thickness. In the present configuration with an anisotropic defect layer, as mentioned above, the polarization state changes after passing through the anisotropic layer depending on the thickness. Then how is the polarization state of the lasing emission?

To study the polarization characteristics of these CLC lasers, we first observed the polarization dependent lasing of a low molecular weight CLC layer. As shown in Fig. 3, L-CP lasing emission with the same handedness as the CLC helix is observed at the low-energy edge of the PBG. A weak peak for R-CP is due to a λ/4 plate, which is not suited exactly to the lasing emission wavelength. This result is consistent with the simulated result [22].

Fig. 4 shows the lasing spectra (solid curve) observed in a cell with a NLC defect layer using R- and L-CP. Here, the reflectance spectrum (dash-dot curve) of PBG region is adjusted to overlap with the fluorescent emission band by controlling the mixing ratio of the chiral polymeric CLC (93 wt%). As shown in Fig. 4, a sharp lasing emission is generated just at the notch of PBG. A remarkable point is that lasing occurs both for L-CP and R-CP. The experiment was also performed in a cell with a decreased amount of chiral polymer NLC (87 wt%) to have a lasing emission at

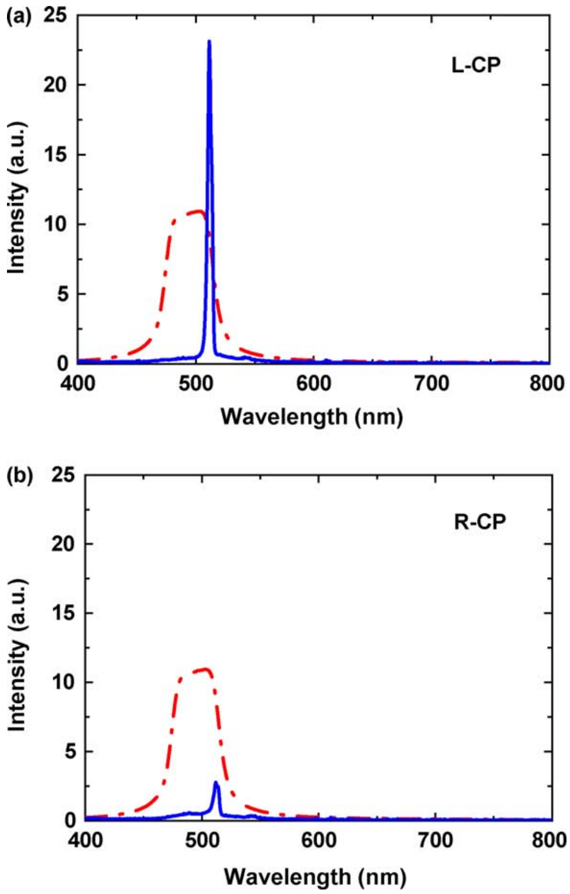

Fig. 3. The L-CP (a) and R-CP (b) lasing emission (solid curve) spectra at the low-energy edge of PBG and reflectance (dash-dot curve) of PBG in a dye-doped single CLC cell.

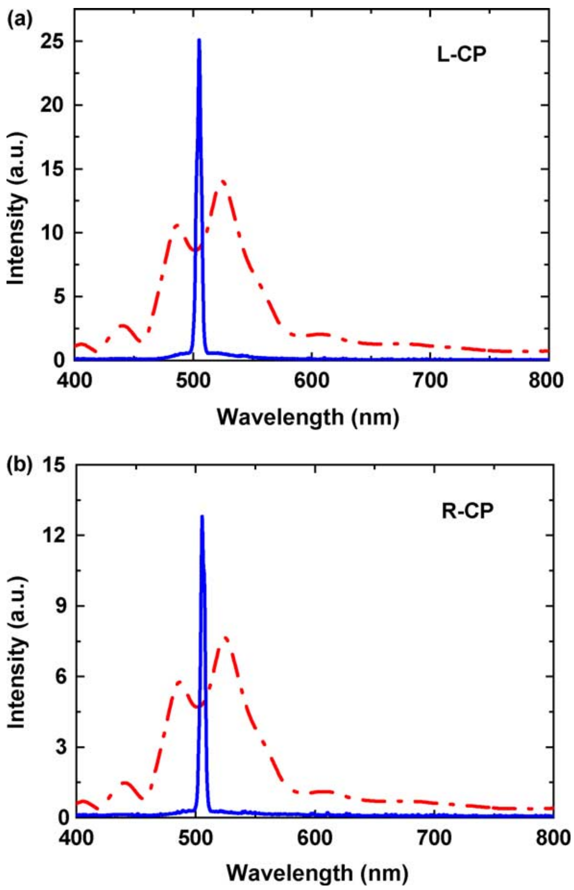

Fig. 4. Lasing emission (solid curve) spectra at the defect mode and reflectance (dash-dot curve) spectra of phase retardation defect mode. (a) L-CP defect mode lasing. (b) R-CP defect mode lasing for the cell with the mixing ratio of chiral NLC polymer of 93 wt% in CLC layers.

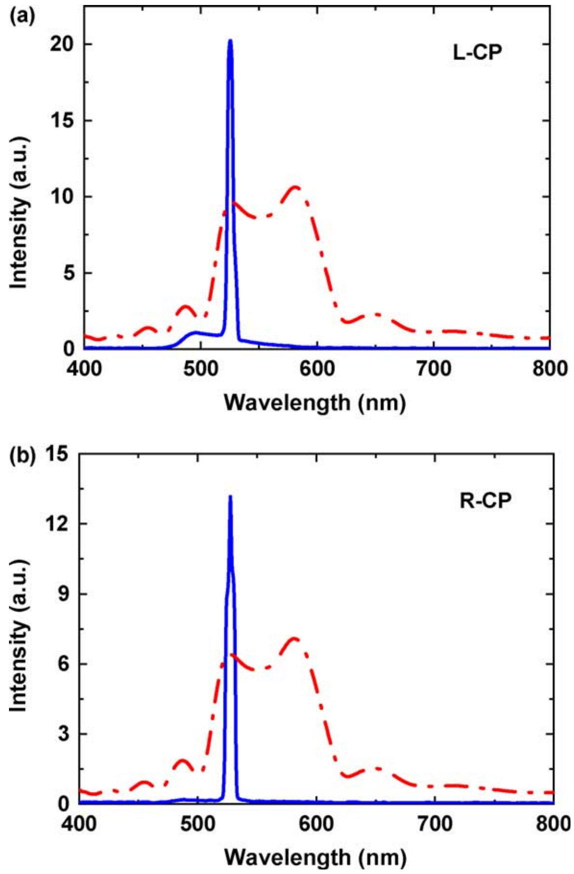

Fig. 5. Lasing emission (solid curve) spectra at the high-energy edge of PBG and reflectance (dash-dot curve) of phase retardation defect mode. (a) L-CP defect mode lasing. (b) R-CP defect mode lasing for the cell with the mixing ratio of chiral NLC polymer of 87 wt% in CLC layers.

the high-energy edge of PBG, as shown in Fig. 5. It is noteworthy that both L-CP and R-CP lasing emissions are observed in this case, although a small spontaneous emission at the higher energy side of the lasing peak can be seen in the L-CP, (Fig. 5(a)) that is absent in the R-CP, (Fig. 5(b)).

4. Conclusions

We have studied the effect of the anisotropic NLC defect layer introduced between PCLC layers, and have been addressed, in particular, the polarization characteristics. It was found that the reflectance was over 50% in the PBG region and both CP lasing would occur irrespective of the modes of lasing, defect mode and edge mode, due to the birefringence of the defect layer.

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