Electro-controllable omni-directional laser emissions from a helical polymeric network composite film

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Keywords : photonic materials, polymeric helical structure, cholesteric liquid crystals, polymeric network, lasing

Abstract

In optical information technology, an electro-controllable Photonic Band Gap (PBG) in a photonic crystal (PC) material is potentially useful for the manipulation of light.[1-4] Despite a great deal of research on PBGs, the reliable use of electro-active PBG material systems is restricted to only a few cases because of the complex and limiting nature of the structures involved. Here, we propose a PBG system that uses a liquid crystal (LC) polymer composite. The composite is made of nematic LCs (NLCs) embedded in polymeric helical networks of photo-polymerized cholesteric LCs (CLCs). The composite film shows a large field-induced reversible color shift over 150 nm of the reflection band, due to the reorientational undulation of the helical axis, similar to the Helfrich effect.

1. Introduction

PCs have been of interest to scientists for over 20 years, because their potential optical properties offer ways of controlling light using PBG materials,[1-4] which preclude the passage of photons within a given energy range. According to physical concepts based on theoretical predictions of PBGs, some promising applications of PCs have been proposed in photonic devices that use three-,[5-7] two-,[8,9] and one-dimensional (1D)[10] PBG material systems. 1D PC materials have many applications, are easy to make, and hence potentially inexpensive. One interesting form of 1D PCs is cholesteric liquid crystals (CLCs) which are characterized by their spontaneous self-assembly into periodic helical structures. CLCs have the unusual optical property of selectively reflecting circular-polarized light; only light that has a different handedness from the CLC helix can propagate in the PBG frequency range. These optical characteristics make the CLCs attractive for many applications, such as mirrorless lasing and reflective color displays, circular polarizers, and color filters. Mirrorless lasing is of particular interest, because the group velocity of the photon approaches zero at the edges of the PBG and thus a low-threshold laser may be possible.

A number of investigations into lasing using CLCs have been reported. Recently, we constructed multilayered structures consisting of CLCs and nematic LCs (NLCs). We succeeded in producing efficient bi-directional lasing by placing an anisotropic defect NLC layer between two CLC layers and in realizing an electro-tunable optical diode with heterojunctions. These are only applicable for forward and backward bi-directional lasing propagations. In this communication, we report on the electro-controllable omnidirectional propagation of laser emissions from a photonic composite film that contains conventional non-reactive NLCs embedded in polymeric helical networks of photo polymerized CLCs. To the best of our knowledge, this is the first time a propagation-tunable lasing device has been

reported using a simple CLC material system.

There are three main ways to control PBGs by applying an electric field in CLC cells, in which the helical axis of the CLCs is normal to the cell surface. These are: (1) by unwinding the helices, (2) by extending / compressing the helical pitches, or (3) by inducing local tilt or the undulation of helices. By using these deformable helices, it is possible to create electrically controllable CLC lasers. Recently, for example, an electricallydeformed helical configuration of a CLC cell was suggested as a new type of wavelengthtunable mirrorless laser device. However, despite the development of such frequencytunable (ω-tunable) lasing devices, the CLC lasing source requires further improvement, as it is still not possible to control the direction of lasing propagation by electrical means (i.e., to construct wavevector-tunable or *k*-tunable lasing devices) using the CLC structures that have so far been proposed. Thus, the realization of fabrication of such simple *k*-tunable lasing devices using a reliable CLC system is an attractive one.

2. Experimental

The investigated Electro-Optic (E-O) cells contain the photonic helical CLC polymer composite films. These films consist of non-reactive NLCs embedded in polymeric helical networks of photo-polymerized cyclic CLC oligomers (Fig. 1a). Before an electric field is applied, the E-O sample cell has a homogeneous planar helical structure, in which the helical axis of the polymer composite is perpendicular to the surface of the substrate. Hence, a welldefined reflection band may be clearly observed from the sample cell. Figure 1b shows the transmission spectra of the planar cells of the composites with several different concentrations of the CLC component. As the concentration of the CLC component increases, the PBG of the composite film clearly shifts to the short wavelength region, while the band-width decreases slightly. The inset reveals a photograph of the sample cells showing distinct selective reflections. Thus, it is clear that homogeneous planar alignments of helical structures were formed over the whole area of the composite cells. The helical pitch and the PBG width of the planar cell as a function of the concentration ratio of the CLC component are also shown in Fig. 1c. As shown in the figure, the twisting power (or inverse pitch) varies in proportion to the concentration of the chiral compound, indicating that these two components (CLC and NLC) are
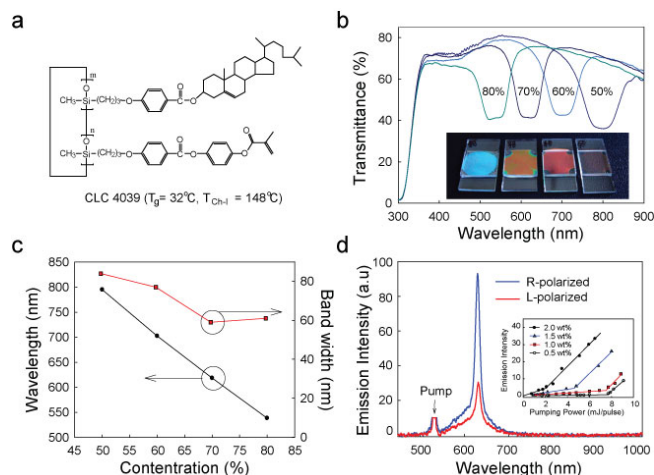

Figure 1. (a) NLCs embedded in polymeric helical networks of photo-polymerized cyclic CLC oligomers (b) Transmission spectra of the planar cells of the composites with several different concentrations of the CLC component. (c) The helical pitch and the PBG width of the planar cell as a function of the concentration ratio of the CLC component (d) The emission peak intensity, plotted against the pumping power, for several concentrations of fluorescent dye.

homogeneously mixed together. It may be seen that after photo-polymerization, the reflection spectra do not change at all, indicating that no phase separation between the chiral and achiral components occurs during polymerization.

In order to confirm the helical structure of the photo-polymerized composite cell, we also measured the emission spectra of the lasing sample cell (CLC concentration: 70 %) which consisted of a dye-doped (dye concentration: 2.0 wt %) helical polymer composite film as an active medium layer, as shown in Fig. 1d. Upon optical pumping at 532 nm, a sharp peak of light emission was clearly observed at the edge of the low-energy band (~630 nm) of the sample cell for right-handed circular polarization (blue curve), which was the same handedness as the helix of the composite. In contrast, a relatively weak emission was observed for left-handed circular polarization (red spectrum). The observed Full Width at Half Maximum (FWHM) of the emission peak is about 3 nm. The emission peak intensity, plotted against the pumping power, for several concentrations of fluorescent dye, is also shown in the inset of Fig. 1d. As may be seen, the lasing threshold decreases as the dye concentration increases: the observed lasing thresholds were about 7.5 mJ/pulse, 7.0 mJ/pulse, 3.9 mJ/pulse, and 1.4mJ/pulse for dye concentrations of 0.5 wt %, 1.0 wt %, 1.5 wt %, and 2.0 wt %, respectively. For

concentrations exceeding 2.0 wt %, aggregation of the dye began to occur, and the lasing behavior became irregular as a result of this inhomogeneity. Thus, the optimum concentration for efficient lasing was around 2.0 wt %. The observed threshold of 1.4 mJ/pulse is fairly low and comparable to the thresholds of similar devices previously reported elsewhere.

3. Results and discussion

We now present the apparent and spectral changes by applying a bipolar square wave of 100 Hz between the pair of substrates (across the E-O sample cell). Color changes from the sample cell, induced by the applied electric field, can clearly be seen. Figure 2a shows the variation of observed transmission spectra from the cell with increased applied voltages (amplitude). From the figure, it is clear that the selective reflection band shifts toward a shorter wavelength region, i.e., from ~700 nm (at 0 V) to ~550 nm (at ~80 V), and becomes broader from ~50 nm to ~160 nm as the applied voltage is increased. Photographs of the sample cell in reflection geometry are also shown in Fig. 2b. From the figure, one can clearly see dramatic field-induced changes in the color of the reflected light from the E-O cell, which shows the electro-tunability of the apparent selective reflection band, even in a CLC polymer network.

In order to better understand this field-induced phenomenon, we also observed the transmission, reflection, fluorescence, and lasing spectra from the cell surface normal, as a function of applied voltages for a fabricated laser sample cell (CLC4039 concentration: 70 %), as shown in Fig. 2c. In these observations, we used a low dye concentration (0.5 wt %) in the sample cell to clearly distinguish the PBG band from the strong optical absorption of DCJTB dye. As shown in the transmission and reflection spectra, a clear blue-shift was also observed for both spectra with increasing external voltage.

The observed electro-tunable blue-shift of the PBG band is quite similar to fieldinduced phenomena such as compression of the helical pitch and local tilt of the helical axis as a result of the Helfrich deformation. In order to understand what is happening here, we also investigated the photoluminescence (PL) spectra and lasing spectra from the laser cell as a function of the voltage applied. In the PL spectra thus obtained, the wavelength region of the fluorescent spectra covers that of the reflection band and as a result, the suppression of the PL spectra due to the PBG band could be observed. From the
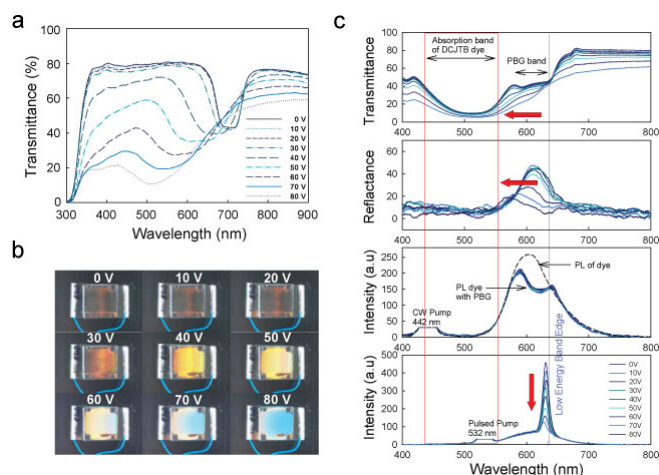

Figure 2. (a) Variation of observed transmission spectra from the cell with increased applied voltages (b) Photographs of the sample cell in reflection geometry are also shown (c) Transmission, reflection, fluorescence, and lasing spectra from the cell surface normal, as a function of applied voltages for a fabricated laser sample cell.

characteristic PL spectra, it is clear that the spectral shape of the fluorescent emission is almost independent of the applied voltage, despite a large blue-shift in the observed selective reflection. In a similar manner to the fluorescent spectra, the peak position of the lasing wavelength at the low energy band edge is also independent of the voltage, although the lasing intensity decreases with increasing applied voltage, as shown in Fig. 2c (bottom). It may be seen that that the real pitch of the composite layer is almost independent of the applied voltage, being distinct from the apparent observation of the field-induced large blue-shift of the PBG band. These results cannot be explained by the previous reported data on controllable PBGs within a CLC structure.

4. Summary

In summary, we investigated laser emissions from an organic laser that has an active medium layer, which consisted of a helical polymer composite film doped with a fluorescent dye. It was demonstrated that efficient circular-polarized laser emissions were produced from the composite film by optical pumping at a relatively low lasing threshold. The wavelength of the emitted laser light corresponds to the low-energy band edge. wave plate and an analyzer to investigate the polarization of the light emitted from the sample.

5. References

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