Effect of Phase Retardation on Defect-Mode Lasing in Polymeric Cholesteric Liquid Crystals**

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Crystals possessing a periodic modulation of their dielectric function are called photonic crystals (PCs), and inhibit certain frequencies of electromagnetic radiation from propagating through specific crystal orientations, (i.e., have a photonic bandgap (PBG)).[1] PCs are widely viewed as promising materials for optical signal processing due to the fact that the flux of the light can be readily manipulated by these materials. Cholesteric liquid crystals (CLCs) are particularly interesting one-dimensional PC materials because of their spontaneous self-assembly into periodic structures and the fact that the PBG can be tuned over a broad range of frequencies. These materials make promising photonic devices possible, making use of lasing at the PBG edge, and examples have been reported in low-molecular-weight CLCs,[2–6] in polymeric cholesteric liquid crystals (PCLCs),[7–9] in chiral smectic LCs,[10] and in a blue phase.[11] Lasing due to physical imperfections introduced into periodic CLCs, i.e., defect modes, has also been reported.[12,13] In the study reported here, we constructed a sophisticated system comprising a polymer-dye-doped nematic defect layer sandwiched between PCLC films and succeeded in introducing the new effect of a retardation defect mode, resulting in modulation of reflectance over 50 % and efficient lasing.

Cholesteric liquid crystals show the unusual optical property of selective reflection of circularly polarized light; that is, light with the same handedness as the CLC helix cannot propagate with a frequency that lies within the PBG.[14] The same phenomenon has also been observed in certain insect cuticles.[15] Even more interesting is a beetle named *Plusiotis resplendens*, which reflects both right- and left-circularly polarized light components.[16] This latter effect originates from a structure consisting of left-handed PBG structure that is bounded on one side by a layer with unidirectional orientation of fibrils that acts as a half-wave ($\lambda/2$) plate. In this manuscript we demonstrate lasing from photonic structures similar to those in the cuticle of *Plusiotis resplendens*.

The discovery that spontaneous emission in fluorescent-dye-doped CLCs can be dramatically influenced by the PBG[17,18] has generated considerable interest in lasing using a distributed feedback (DFB) cavity that is formed spontaneously in the CLC. Polymerized CLC films offer reduced lasing threshold, high quantum yields, improved emission,[7] and good processability, and are good organic material candidates for continuous wave lasing and electrically pumped organic diode lasers, both of which have many practical applications. Polymeric CLC films (PCLCs) can be used for the formation of a Fabry–Pérot etalon and hence can be extended to produce tunable filters.[19]

The use of defect modes inside PBGs[20] has also been proposed for generating lasing at low thresholds.[20,21] Generally, two kinds of configurations are theoretically suggested to generate a defect mode in CLCs: the introduction of an isotropic spacing layer in the middle of the CLCs,[22] and the creation of a phase jump without a spacing layer in CLC[23,24] and smectic LC structures.[25] Oldano et al. examined the propagation of shear waves instead of light.[24] Recently, defect-mode lasing has been experimentally demonstrated by using a phase jump in CLCs[12] and by electrically tuning the wavelength of the defect mode with a layer of nematic liquid crystal (NLC) inserted into dielectric multilayers.[13] However, reports of defect mode lasing from structures that include both a spacing layer defect and the phase effect have not yet appeared.

To ensure that the molecular orientation (see Fig. 1a) at the air surface is actually parallel to the surface, we first fabricated a nematic cell sandwiched between polymer NLC films and a conventional NLC cell between substrates with polyimide surface alignment layer, and measured transmittance for the cells between crossed polarizers as a function of sample rotation angle. Both samples show four peaks with zero transmittance minima for 360° rotation. This ensures that the polymer LC film acts as an alignment layer, meaning that the air-surface of the polymer LC also shows good planar alignment. This may originate from the polymer LC, being an advantage of using a polymer. Next the same transmittance measurements as above were carried out using two cells, i.e., a dye-doped nematic layer sandwiched between photocurable CLCs

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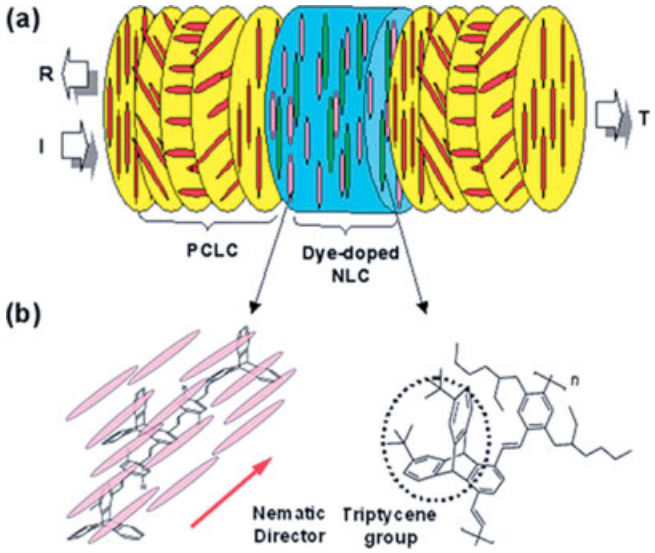

Figure 1. a) Schematic illustration of the cell structure with a polymer-dye-doped NLC defect layer sandwiched between PCLC films. The thickness of PCLC films is 1.8 μm each, and that of the defect layer is 2 μm. b) The chemical structure of the polymer dye used (T-PPV). The main chain of the polymer-dye is well aligned along the local director of the PCLC, and the triptycene group (dotted circle) suppresses orientation fluctuation of LC molecules, resulting in a higher order parameter.

and a dye-doped nematic layer sandwiched between our PCLCs, to confirm parallel director orientation at both surfaces of PCLC films by comparing the result with that of photocurable CLCs that have well-defined director orientations at surfaces as a reference. Thus we confirmed the cell structure shown in Figure 1a.

As shown in a dashed circle in Figure 1b, the triptycene groups of the fluorescent polymeric dye triptycene poly(*p*-phenylenevinylene), T-PPV, suppress the fluctuation of LC molecules and prevent the strong interpolymer interactions often associated with semiconducting polymers that generally result in low solubility and emission efficiency. As a result, the triptycene provides highly emissive compositions[26–28] in which the main chains (emission transition moment) are highly ordered along the local director of the NLC host. We have shown previously that this arrangement produces low-threshold and efficient single-mode lasing.[5,6]

Reflectance spectra were acquired using a microscope spectrometer (TFM-120AFT-PC, ORC). For fluorescence and lasing measurements, a 440 nm pulsed laser beam from an optical parametric oscillator (Surelite OPO; HOYA Continuum) pumped by third-harmonic light from a Nd:YAG (yttrium aluminum garnet) laser was used as the 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 along the direction parallel to the helical axis (normal to the substrate) and then detected by a multichannel spectrometer (USB 2000: Ocean Optics, Inc).

The measured reflectance spectrum from a fabricated sample cell with a defect layer of 2 μm thickness is shown as a red curve in Figure 2a. For comparison, the reflectance spectrum

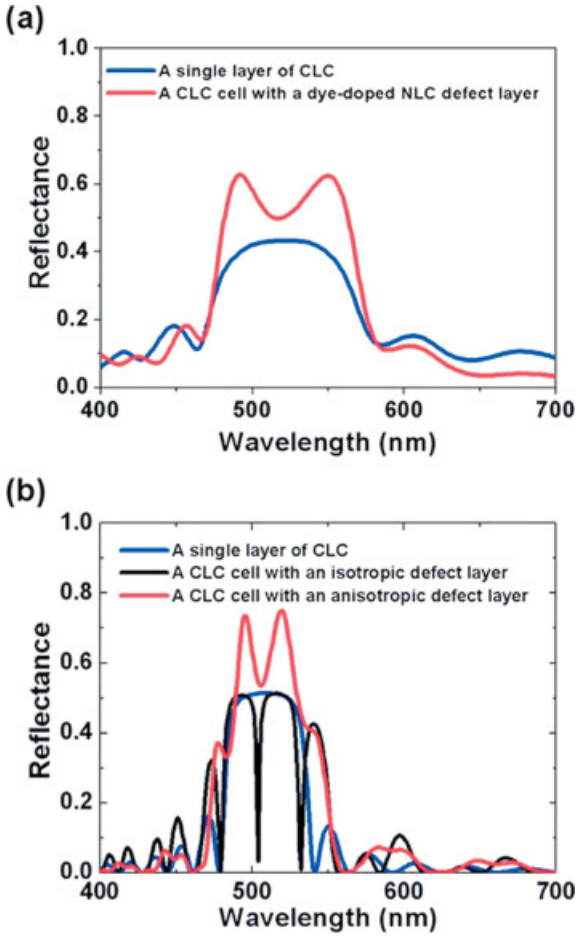

Figure 2. a) Reflectance spectra from a single layer of PCLC (blue curve) and a PCLC cell with a T-PPV-doped NLC defect (red curve). The PCLC with a defect layer shows reflectance greater than 50%, while the reflectance of the PCLC without a defect layer is about 40%. b) Simulated reflectance spectra of a single layer of CLC (blue curve), a CLC cell with an anisotropic defect layer (red curve), and a CLC cell with an isotropic defect layer (black curve). The cell geometry and materials parameters for the simulation are as follows: The defect layer of 1.92 μm thickness is sandwiched between PCLCs 1.78 μm thick. The CLCs have refractive indices $n_e = 1.63$ and $n_o = 1.5$ and optical pitch 510 nm. The indices of the anisotropic layers are $n_e = 1.66$ and $n_o = 1.5$, and the indices of the isotropic layers are $n_e = n_o = 1.55$. For the anisotropic defect layer, the above parameters give the condition that the directors at both surfaces of PCLCs are parallel to each other, as is the nematic director in the defect layer. The pretilt of the anisotropic LC layer is considered as 0° for simplicity.

of a single PCLC film without a defect layer is also shown, as a blue curve, in the figure. Two important observations from the data in Figure 2a are that 1) a notch caused by a defect layer is observed in the red curve, and 2) the reflectance is greater than 50 % in the cell with a defect layer, while it is less than 40 % in the cell without a defect layer.

The reflection originates from selective reflection, in which one circularly polarized light component with the handedness the same as that of the CLC helix is totally reflected, but the other circularly polarized component is transmitted without reflection. Therefore the reflectance essentially cannot exceed

50 % in a single CLC cell, as shown by the blue curve in Figure 2a, where the lower reflectance is due to the thin nature of PCLC films. Hence the observed reflectance above 50 % is a striking new phenomenon that exceeds those of previously suggested defect modes in CLCs. This modulation of reflectance over 50 % is caused by the phase retardation of the birefringent defect layer. To understand this better, let us consider an extreme case such as a cuticle of *Plusiotis resplendens*, in which a defect layer acts as a $\lambda/2$ plate. The circular polarization with the opposite handedness to the PCLC helix is transmitted through the first CLC layer, but then changes its polarization state to the opposite circular polarization, which is selectively reflected by the second CLC layer. This reflection directs the light back through the $\lambda/2$ plate defect layer, where the polarization again returns to its original transmitted state, to give reflection greater than 50 %.

To confirm that the optics principles mentioned above are operative, reflectance spectra in three types of cells were simulated. Namely, calculations were performed for cells 1) without a defect layer, 2) with an isotropic defect layer, and 3) with an anisotropic (nematic) defect layer. The parameters used for the calculations are shown in the figure legend. As clearly shown in Figure 2b, only the cell with an anisotropic defect layer displays a maximum reflectance above 50 %; the other two cells—without an anisotropic defect layer or with only an isotropic defect layer—show reflectance less than 50 %. The shape of the notch in the two types of defect modes is also characteristic. The isotropic defect layer displays a sharp notch, while the anisotropic defect layer produces a broad notch. In this way, we theoretically observed a defect mode that is influenced by the phase retardation.

Even though the notch resulting from the defect is not sharp, the reflectance is high, providing advantages for lasing action for the following reason. In the usual single-CLC laser system, it is difficult to obtain relatively high gain with low threshold for lasing, because the photonic bandgap is defined only for the circularly polarized light of one handedness. This means that nearly half of the emission from the excited molecules cannot be incorporated in the lasing action. On the other hand, in our system, the high reflectance results from the changes in the polarization state on passing through the anisotropic layer. This indicates that circularly polarized light of both types of handedness feels the photonic band. In fact, the photonic bandgap appears for both right- and left-circularly polarized light. Hence, lasing action for both circular polarizations is possible and one can obtain relatively high gain. This is actually the case, as will be shown below. The advantage of the present lasing action is that the magnitude and shape of reflectance can be controlled by adjusting the thickness and the refractive index of the anisotropic NLC defect layer.

In our designs the defect layers have profound effects on the fluorescence and lasing behavior of T-PPV. Figure 3 shows

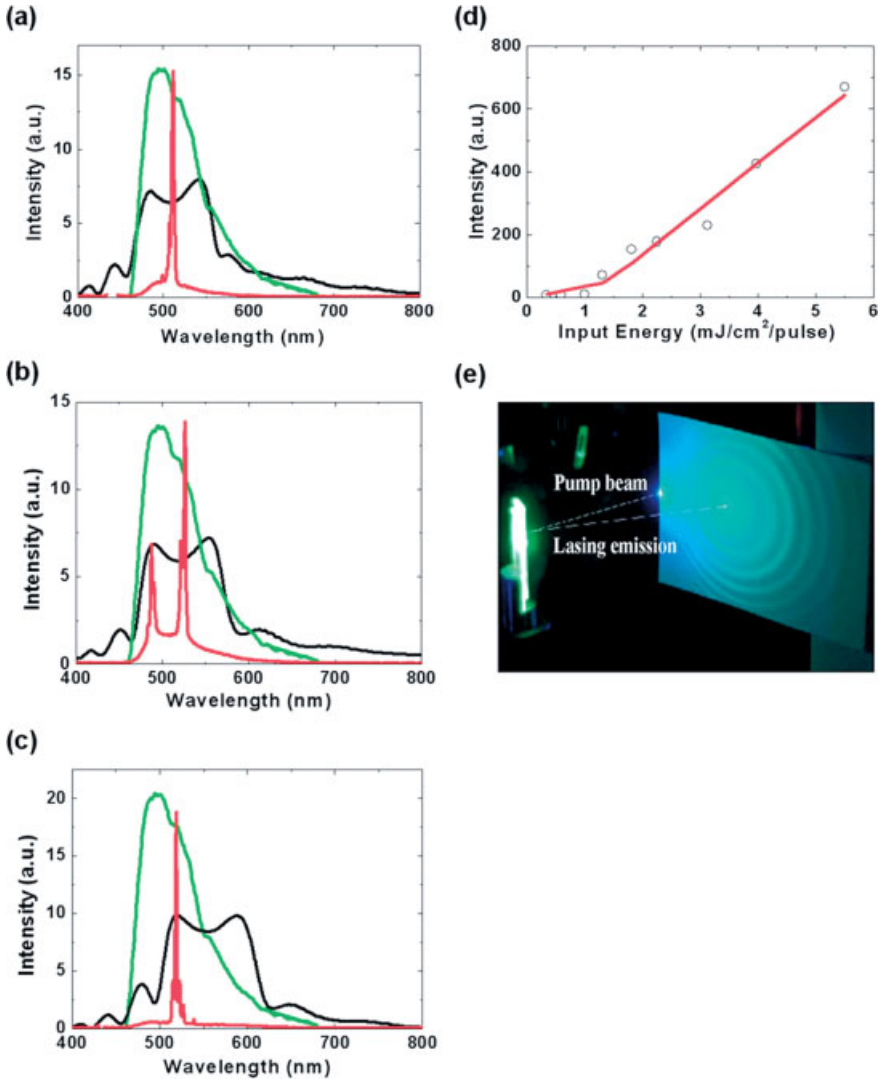

Figure 3. Fluorescence (green curve), reflectance (black curve), and lasing emission (red curve) spectra for three cells with a defect layer 2 μm thick and with different mixing ratios of the component with chiral units: a) 93 wt.-%, b) 92 wt.-%, and c) 87 wt.-%. The chiral content specifies the position of the PBG with respect to the emission band. Lasing occurs a) at the defect mode, b) at both the defect mode and the PBG edge, and c) at the PBG edge. d) Threshold behavior in the defect mode. e) Photograph of a far-field pattern of the laser emission using the defect mode. Concentric rings are indicative of good optical coherence.

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the results in three cells with nematic defect layers of the same thickness (2 μm) and with different mixing ratios of the component with chiral units: a) 93 wt.-%, b) 92 wt.-%, and c) 87 wt.-%. The different chiral contents adjust the position of the reflection band (black curve) with respect to the emission band (green curve).

In the first example, shown in Figure 3a, the reflectance spectrum was adjusted to overlap with the fluorescent emission band. The wavelength of the defect-induced notch (~500 nm) is coincident with the fluorescence peak of T-PPV. This situation promotes lasing activity, as shown by the red curve. The defect mode lasing appears as a Lorentzian peak at 511 nm with full width at half maximum (FWHM) of about 3 nm. It should be noted that in this case the sample cell is considerably thinner (5.6 μm) than has been previously reported for defect mode lasing (34 μm).[12] This result demonstrates the high efficiency of anisotropic defect mode lasing and that this approach has key differences from the conventional defect mode lasing in CLCs.

The second example is a cell of PCLCs with a NLC defect layer and a PBG reflectance spectrum adjusted to deviate slightly from the fluorescent emission band, as shown in Figure 3b. For this condition, two lasing modes are possible and in addition to the defect mode lasing emission also occurs at the high-energy edge of PBG. The lasing intensity at the defect mode is more intense than that at the high-energy PBG edge mode, thereby indicating that lasing at the defect mode is more efficient. In fact, on increasing the pumping power, defect mode lasing occurred first and then high-energy band-edge lasing occurred later.

The third example is a cell of PCLCs with a NLC defect layer and reflectance spectrum of PBG adjusted to deviate further from the fluorescent emission band, as shown in Figure 3c. For this condition, only one sharp lasing mode is observed, at the high-energy PBG edge. We confirmed that both left- and right-handed circularly polarized band-edge laser lines can be observed.[29] Although only lasing at the high-energy edge of the PBG is observed, the total thickness of the sample cell (about 5.6 μm) is much thinner than those of conventional CLC lasers (typically 15–30 μm).[2,3,5–7,9] This result also indicates that our PCLC laser configuration with a dye-doped NLC layer provides a highly efficient lasing condition in comparison with conventional dye-doped CLC mirrorless laser assemblies.[5,6] In the present case, the high-energy band edge coincides with the emission peak wavelength, so that lasing due to the photonic edge occurs at the high-energy band edge. If the low-energy band edge coincided with the emission peak wavelength, we would expect lasing at the low-energy band edge.

Figure 3d shows the threshold behavior for the defect-mode lasing. The threshold was found to be 1.3 mJ cm^{-2} per pulse. Next, in order to unequivocally confirm the lasing at the defect mode shown in Figure 3a, we observed the lasing far-field pattern. As shown in Figure 3e, simple inspection reveals a clear ring pattern, indicating coherent laser emission at the defect mode. It is also important to note that we confirmed that the lasing emission from the defect mode has both right- and left-circularly polarized components.[29] This fact clearly establishes the role of the birefringent layer.

In this way, we showed the first example of lasing using an anisotropic defect layer. We want to emphasize the difference from those using isotropic defect layers and distributed Bragg reflection (DBR) structures in many respects, such as polarization selectivity, active layer thickness, and so on, which enable us to achieve lasing due to not only the defect mode but also the band edge mode. We also found an interesting lasing action using a very thick (100 μm) anisotropic defect layer. The details will be reported soon.

Finally, we want to make a brief comment from the theoretical point of view. We simulated the density of states and transition probability, the product of which gives a measure of the emission intensity, as a function of wavelength for cells with isotropic and anisotropic defect layers. We found that an anisotropic defect structure is more efficient for lasing than the isotropic defect structure. The details will also be reported in the future.

In conclusion, we have studied the effects of an anisotropic NLC defect layer introduced between PCLC layers. It was found that the modulation of reflectance exceeds the 50 % limitation provided by simple CLC PBGs due to the birefringence of the defect layer. Moreover, by adjusting the PBG region to be coincident with the fluorescent emission band for the guest polymer dye, sharp defect-mode lasing was observed successfully at the defect mode in the middle of PBG. From these results, it was demonstrated that the configuration of a PCLC with a polymer-dye-doped anisotropic defect layer is very attractive for lasing.

Experimental

Mixtures of two NLC aromatic polyester polymers (Nippon Oil Corporation) were used to make the polymeric CLCs. One of the NLC polymers contains 25 % chiral units in its chemical composition. By changing the ratio of the two NLC polymers in the final composition, the wavelength of the PCLC's PBG was controlled. The PCLC films were fabricated by a spin-casting procedure on glass substrates coated with unidirectionally rubbed polyimide (AL 1254, JSR). The PCLC coatings were cured for 2 min at 180 °C, and yielded well-aligned films. The helical axis of the approximately 1.8 μm PCLC film was normal to the substrate surface and two PCLC coated substrates were stacked face-to-face separated by spacers and sealed with the directors at two surfaces parallel to produce a vacant cell. Dye-doped NLCs were prepared by mixing a commercial NLC (ZLI2293, Merck) and a chloroform solution of triptycene PPV (T-PPV), whose precise chemical structure was shown in [28] (see also Fig. 1). After evaporation of the chloroform at 70 °C, the concentration of T-PPV in the NLCs is 2 wt.-%. The dye-doped NLCs were then introduced into the empty cell formed from the PCLC films using capillary action to produce an anisotropic defect layer as shown in Figure 1a.

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Organic Polarized Light-Emitting Diodes via Förster Energy Transfer Using Monodisperse Conjugated Oligomers**

By *Andrew C. A. Chen*, *Sean W. Culligan*, *Yanhou Geng*, *Shaw H. Chen*,* *Kevin P. Klubek*, *Kathleen M. Vaeth*, and *Ching W. Tang*

Conjugated polymers are promising for electronic and photonic applications, such as thin-film transistors, light-emitting diodes, photovoltaics, and sensors.[1,2] In particular, blue organic light-emitting diodes (OLEDs) have been demonstrated using polyfluorenes.[3–5] Emission of green and red light has also been demonstrated via modulation of electronic energy levels through copolymerization.[3,6–11] Linearly polarized OLEDs are potentially useful as an efficient light source for liquid-crystal displays (which account for the bulk of flat-panel displays today), for electroluminescent displays with reduced glare and increased contrast, for projection displays, and for stereoscopic imaging systems.[12,13] The concept of linearly polarized electroluminescence, except in the case of white light, has been demonstrated by taking advantage of the thermotropic nematic mesomorphism exhibited by polyfluorenes and other conjugated polymers.[4,5,14–26] The idea is to preserve uniaxial molecular alignment in a solid film upon thermal annealing, with subsequent cooling through the glass-transition temperature without encountering crystallization. From the materials perspective, monodisperse conjugated oligomers offer numerous advantages over polydisperse conjugated polymers. Monodisperse conjugated oligomers are characterized by a well-defined and uniform molecular structure as well as superior chemical purity via recrystallization or column chromatography. In the absence of chain entanglements or defects (e.g., bends and kinks), relatively short and

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