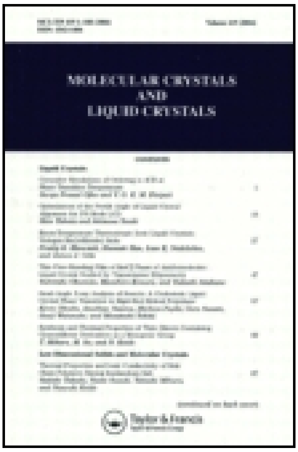

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Fast and Continuous Tunable Lasing from a Nano-Pore Embedded Cholesteric Liquid Crystal Film

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Continuous tuning of lasing wavelength is demonstrated using a photopolymerized cholesteric liquid crystal film embedded with nano-sized liquid crystal droplets. The film is fabricated simply by irradiating ultraviolet light on a mixture comprising photopolymerizable and non-photopolymerizable cholesteric liquid crystals. Because the droplets are smaller than the wavelength of light, they do not contribute to scattering but only to the effective refractive index of the composite: tuning of the lasing wavelength is achieved by applying a field to reorient the liquid crystal molecules in the droplets, thus changing its refractive index. These electro-tunable films also show fast tuning properties and improved thermal stability, making them potentially useful to realize ultra-compact, tunable dye lasers.

Keywords Cholesteric liquid crystal laser; electrically tunable laser; polymerized liquid crystals; refractive index tuning

Introduction

Cholesteric liquid crystals (ChLCs) are known to self-organize into helical structures with periodicities of a few hundred nanometers and thus possess a reflection band $\Delta\lambda$ ($= \Delta n p$) attributed to their periodicity p and birefringence Δn. By incorporating appropriate gain materials (usually organic dyes) in ChLCs, lasing can be realized by optical pumping [1]. ChLCs possess several properties which make them attractive as lasers: they include (1) intrinsic single-mode lasing at the band-edge wavelength of the selective reflection band, owing to the periodic modulation of the dielectric tensor; (2) circularly polarized lasing due to their helical structure; (3) rich diversity in possible methods of wavelength tuning; and (4) spontaneous structure formation, making them cost-effective. ChLC lasers are being pursued as realistic alternatives to conventional dye lasers based on isotropic dye solutions.

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Lasing from ChLC occurs at the band-edge wavelength of the selective reflection band because of the enhancement in the photon density of states at these wavelengths. Tuning of ChLC lasers has therefore been achieved by shifting the selective reflection band by external stimuli such as heat [2,3], electric field [4–7], or light [8,9]: however, in almost all of the tuning methods proposed to date, tuning has been achieved by changing the pitch of the helix. Although pitch modulation allows a large tuning range to be attained, the speed of changing the macroscopic structure is slow, usually on the order of seconds, and the shift is discrete, on the order of several nanometers, because the molecules are usually strongly anchored at the substrate surface, only allowing discrete shifts in the number of helical turns in the LC cell [3]. Hybrid structures where LCs are embedded in helical polymer networks lead to fast response times on order of microseconds [10]. However, the fabrication procedure of such structures are often complex (requiring rinsing and refilling of the cell) and consequently, the quality of the selective reflection band deteriorates, i.e., broadened bands with decreased reflectivity are often observed, deteriorating also their lasing performance.

A truly (non-discretely) tunable laser with a fast response is thus still a challenge in ChLCs. Herein, we report the fabrication of ChLC laser that can be tuned continuously with sub-millisecond response. Our approach is based on modulating the refractive index to control the selective reflection band, while keeping the helical structure of the ChLC fixed. We show that this mode of wavelength tuning can be realized by fabricating a photopolymerized ChLC film embedded with nano-sized pores filled with LCs that can respond to the applied field.

2. Device Fabrication and Experimental Methods

Our sample was prepared by mixing three LC materials from Merck, 03-008 (photopolymerizable ChLC mixture), BL-011 (nematic LC mixture) and R-811 (chiral dopant) at a weight ratio of 03-008: BL-011: R-811 = 46.8: 46.8: 6.4. The chiral dopant was added in the mixture to compensate for the decrease in the helical twisting power of 03-008 and to make the selective reflection band appear in the visible wavelength region. As the gain medium, a laser dye (Pyrromethene 597, purchased from Exciton) was doped in the sample at a concentration of 1 wt%. The ChLC laser was fabricated by injecting the sample in a 10 μm-thick indium tin oxide (ITO) coated sandwich cell with planar rubbing treatment (purchased from E.H.C Co.), and photopolymerizing the mixture by irradiating relatively strong UV light (Yamashita-Denso, Hypercure 2000) at a power of 100 mW/cm^2. To investigate the morphology of the film, the film was rinsed in supercritical CO$_2$ (Rexxam Co. Ltd. SCRD401), and observed by a field-emission scanning electron microscope (SEM, Hitachi S-4300).

The lasing characteristics was investigated by measuring the output upon optically pumping the ChLC film with a pulsed laser. The pump source was a diode pumped, passively Q-switched, frequency-doubled Nd-doped yttrium aluminum garnet laser (CryLas FDSS532-150) with a pulse width of 1.5 ns and repetition rate of 10.00 Hz. The excitation laser beam was focused over a circular area with a spot diameter of approximately 20 μm. The emission spectrum from the ChLC was monitored in the cell-normal direction by using a CCD spectrometer (Ocean Optics, USB2000) with a spectral resolution of 1 nm. High-speed acquisition of the lasing spectra was performed using a streak scope (Hamamatsu C4334) with a time resolution of 15 picoseconds.

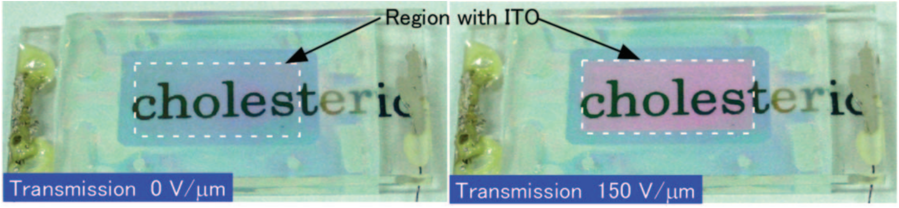

Figure 1. Photograph of the nano-pore embedded ChLC film.

3. Results and Discussion

Figure 1 shows a photograph of the ChLC film, taken before and after application of an electric field. When the electric field was applied, the color of the film clearly changed, and returned to its original color when the field was removed. Field-tuning of the reflection band had excellent reproducibility (stable for at least few million times) and the sample maintained high transparency at all applied voltages, even though it contained 50 wt% of non-photopolymerizable ChLC. The reflectance spectra of the sample shown in Fig. 2 also supports this observation, showing neither broadening nor decrease in reflectivity even at high applied voltages.

To determine the origin of the reflection band shift, the sample was observed under a polarized optical microscope while applying voltage. As shown in Fig. 3, the Grandjean lines found near the edge of the cell did not move, even though the color of the film changed. Since the Grandjean lines are present where there is a discontinuous change in the number of helical turns, the fact that they remained stationary indicates that the helical pitch was not affected by the application of field. Rather, SEM observations indicate that the shift in reflection band was caused by a change in the effective refractive index of the film. The SEM image taken after supercritical CO_2 rinsing (Fig. 4) reveals the existence of small pores several tens of nanometers in size, embedded in the polymer matrix. In a conventional polymer dispersed LC which utilizes a non-liquid crystalline polymer, LC droplets with micrometer order are formed inside the polymer matrix, because of phase-segregation. The nano-sized droplets observed here suggest that only minute segregation occurred in our

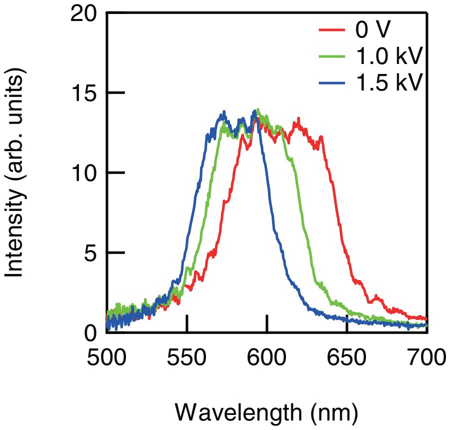

Figure 2. (a) Reflection spectra of the ChLC film at various electric fields.

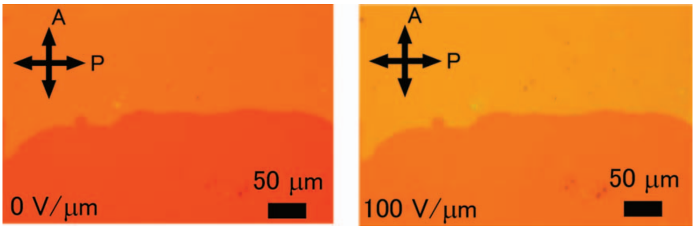

Figure 3. Polarized optical micrographs of the sample before and after applying field.

sample: we believe this is because both the polymer and LC possessed helical ordering (leading to higher miscibility) and because polymerization occurred quickly, using intense UV light. Because the diameter of the pores are smaller than 1/10 of wavelength of light, they do not contribute to scattering, but to the effective refractive index. Thus, by electrically driving the LC molecules in these pores, the effective refractive index of the film can be tuned. The NLC BL-011 used in this study has positive dielectric anisotropy, meaning that the molecules orient along the electrical field, causing the effective refractive index to decrease. The decrease in the refractive index is consistent with the direction of shift of the selective reflection band upon application of an electric field.

Figure 5 shows the tuning characteristics of lasing from the ChLC film, measured as a sinusoidal electric field (150 V/μm) with frequency of 600.0 Hz was applied on the film. Single-mode lasing was observed from the longer band-edge wavelength due to distributed feedback (DFB) in the ChLC. As the phase between the pump (10.00 Hz) and modulation voltage (600.0 Hz) was varied, the lasing wavelength was seen to shift continuously, following a sinusoidal profile (inset, Fig. 5). The sinusoidal dependence of the lasing wavelength on the phase difference indicates that the film is actually being modulated sinusoidally at 600.0 Hz following the voltage signal, although lasing seemed to occur at a single wavelength because pumping was synchronized to the modulation voltage.

Conventional nematic LCs and ChLCs in which elastic interactions are dominant cannot respond to electrical fields this fast. The fast response in this film was made possible by the local reorientation process which occurs only within the nano-pores. To investigate

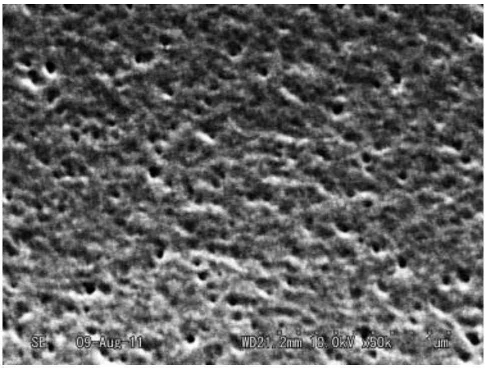

Figure 4. SEM image taken after rising the sample with supercritical CO2.

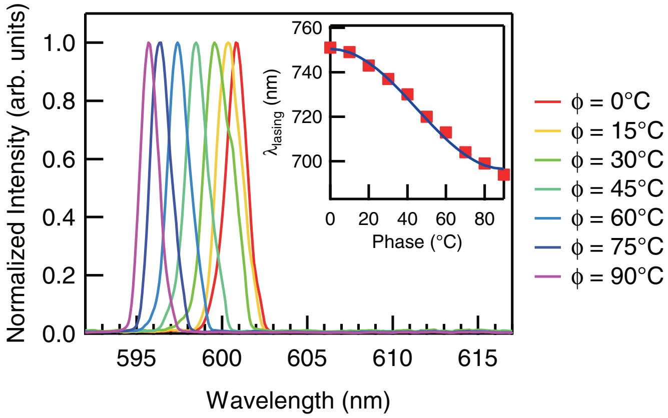

Figure 5. Phase dependence of the lasing wavelength of nano-pore embedded ChLC laser.

the tuning properties further, lasing spectra were measured while tuning the sample at 200 Hz and pumping at 1 kHz, which is the limit of our pump laser. It can be seen from Fig. 6 that each shot occurring at 1 millisecond interval is emitted at a different wavelength, thus proving the fast tuning capability of the ChLC laser.

Our device has another advantage in terms of thermal stability of lasing wavelength. High sensitivity of the pitch to thermal environment and resultant drift of lasing wavelength, reaching few 10 nm/°C is one of the major drawbacks of ChLC lasers [11]. By fixing the helical structure by photopolymerization, the temperature stability can be improved significantly. Shown in Fig. 7 is the temperature dependence of the lasing wavelength measured without field. The lasing wavelength varies very slowly with temperature, at a rate of −0.3 nm/°C between 30 and 70°C. Above 75°C at which the LC in the nano-pores melts to the isotropic phase, the stability increases even further to -0.008 nm/°C. The tiny shift in wavelength by temperature is important for long-term stability of ChLC lasers in practical applications.

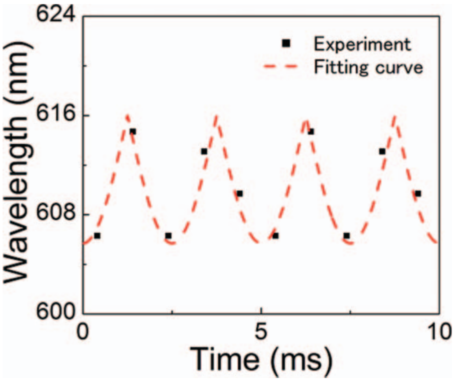

Figure 6. Lasing wavelength of the sample at pump frequency of 1 kHz and modulation frequency of 200 Hz.

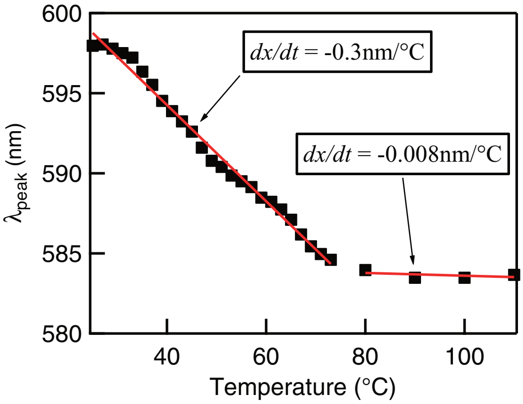

Figure 7. Temperature dependence of the lasing wavelength of the sample.

5. Conclusions

Continuous tunable lasing was demonstrated using a nano-pore embedded ChLC film, which changes its effective refractive index by applying an electrical field. The film also showed fast response, improved thermal stability and excellent reproducibility. Because of the simple fabrication procedure and flexibility, these nano-composite films are potentially useful to realize ultra-compact wavelength-tunable dye lasers.

Acknowledgments

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