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Electric Field Dependence of Lasing Wavelength in Cholesteric Liquid Crystal with an In-Plane Helix Alignment

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The authors report continue tuning of the lasing wavelength in cholesteric liquid crystals (ChLCs) by an applied electrical fields. Although the pitch length and the corresponding lasing wavelength is sensible to external fields, only a discrete shift in the pitch is observed in the conventional cell configuration where the helix axis is lying the cell-normal direction, since the molecules are anchored to a single direction at the substrate interfaces. In this study, continuous wavelength tuning is achieved by laying the helix in the cell-plane direction, mitigating the effects of anchoring: this alignment is attained simply by applying an electric field while cooling the sample from the isotropic state, in a planarly rubbed sandwich cell. We also show that the tuning characteristics as that predicted based on calculations using the elastic theory.

Keywords Cholesteric liquid crystal; in-plane helix alignment; lasing action

1. Introduction

Cholesteric liquid crystals (ChLCs) are known to self-organize into helical structures about a single helix axis. In such periodic birefringent media, a phenomenon called selective reflection occurs: light falling within the wavelength region $\lambda = n_o p - n_e p$ where n_o, n_e and p are the ordinary and extraordinary refractive indices and helical pitch, respectively, is reflected because of Bragg reflection. Furthermore, only light with the same circular handedness as the material is reflected because of the strongly twisting structure. The sensitivity of the pitch to external fields has made ChLCs an attractive material as tunable reflectors and polarization elements [1,2].

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Laser action in ChLCs is also a subject that has attracted a lot of scientific and practical interest [3–10]. Light at the wavelength of the selective reflection band edge forms a standing wave and its group velocity is suppressed within the ChLC. For this reason, stimulated emission is encouraged if there is an active medium doped in the material. Indeed, single wavelength lasing can be obtained from the band-edge wavelength by optically pumping a ChLC doped with an appropriate dye. Furthermore, the lasing wavelength can generally be tuned over a broad spectrum range because of the pitch tenability of the ChLC. Consequently, the ChLC laser has attracted a large amount of interest as small-size, low-threshold and tunable laser [3–10].

Previous works have shown that in a conventional ChLC laser with a planar alignment (where the helical axis is perpendicular to the substrates), only a discontinuous shift of lasing wavelength is observed because strong molecular anchoring at the substrate surfaces: both winding and unwinding of the helix must be accompanied by a $\pm\pi$ turn of the anchored molecules [11]. There are two approaches to overcome this problem: one is to increase the thickness of the cell to decrease the jump in the pitch change; and the other approach is to align the ChLC in such a way that the helix is lying in the cell-plane direction to mitigate the effects of anchoring on the elongation or shortening of the helix [1,12,13]. In this study, we utilize the latter approach, report continuous tuning of the lasing wavelength by an applied electric field. We will first show the experimental procedure to attain the in-plane helix alignment, then its laser emission characteristics. We also show that the tuning properties agree well with the theoretical prediction obtained from elastic theory [14–18].

2. Experimental Setup

A left-handed ChLC was prepared by doping a chiral dopant (Merck, S-811) and of a laser dye, [2-[2–4(dimethylamino)pheny1]etheny1]-6-methy1-4H-pyran-4-ylidene propanedinitrile (Exciton, DCM) at 32 wt% and 0.5 wt% respectively, in a nematic LC host (Merck, E44). The clearing point of the material was approximately 68°C and its refractive indices were $n_e = 1.72$ and $n_o = 1.54$ respectively. The ChLC was infiltrated between a 6 μm-thick sandwich cell composed of In-Sn-oxide (ITO) coated glass substrates of which the surface was spin coated with polyimide (JSR, AL1254) and rubbed in a single direction. The rubbing direction of the top and bottom substrates of the assembled cell was parallel.

In order to attain the in-plane helix alignment, the cell was cooled from 73.0°C, which is about the clearing point of the ChLC, to room temperature at 0.5°C/min while applying an electric field. A rectangular-wave electric field was applied with a function generator (Hewlett Packard, 3314A) and an amplifier (NF Electronics Instruments, 4010); its amplitude and frequency were 2.7 V/μm and 1 kHz, respectively. The texture of the cell was observed by a polarization optical microscope (POM) (Nikon, multiphoto2-pol) and the direction in which the ChLC helix axis lied was determined using a Berek compensator.

Figure 1 shows the experimental setup to measure the emission spectra. The second harmonic light of a Q-switched Nd:YAG laser (Spectron Lasers, SL802) was used to excitate the dye doped in the ChLC; the wavelength, pulse width, and pulse repetition frequency were 532 nm, 20 ns, and 10 Hz, respectively. The excitation laser beam was focused by a cylindrical lens to an area of approximately 0.1 mm^2, and was irradiated on the sample so that the long axis would be parallel to the helix

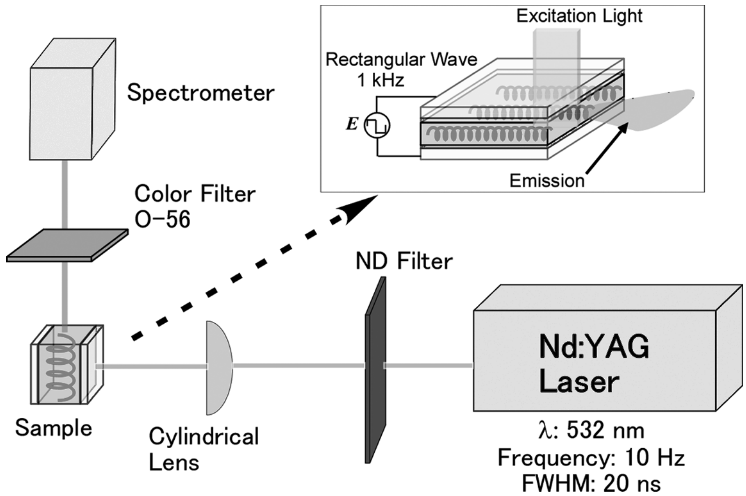

Figure 1. Experimental setup for emission spectrum measurement.

axis. The outgoing emission in the transverse direction from the cell was measured using a multichannel spectrometer (Hamamatsu Photonics, PMA-11) with a spectral resolution of 2 nm.

3. Result and Discussions

The alignment of the CLC was investigated on the POM by rotating the cell at 35.0°C. Figure 2 shows the POM images taken with the rubbing direction either at an angle of 45° or 0° to the analyzer. The intensity of the transmitted light changed gradually upon rotating the cell and a completely dark state was obtained at 0° to the analyzer, indicating that the texture is optically uniaxial with the optical axis either parallel or perpendicular to the analyzer. This also means that the ChLC is aligned with a uniformly lying helix [20], and the direction in which the helix axis lies corresponds to the direction of the optical axis. The optical axis was determined using a Berek compensator and was found to be parallel to the rubbing direction. This alignment is believed to be a result of the balance between two forces: that is, the molecules anchored at the substrates surface gradually twisting into the bulk, and the molecules in the bulk experiencing a torque to orient themselves in the cell-normal

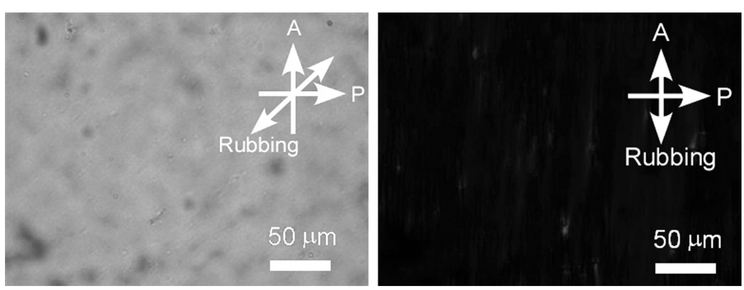

Figure 2. Polarizing microscope images of CLC at an angle of 45° or 0° to an analyzer.

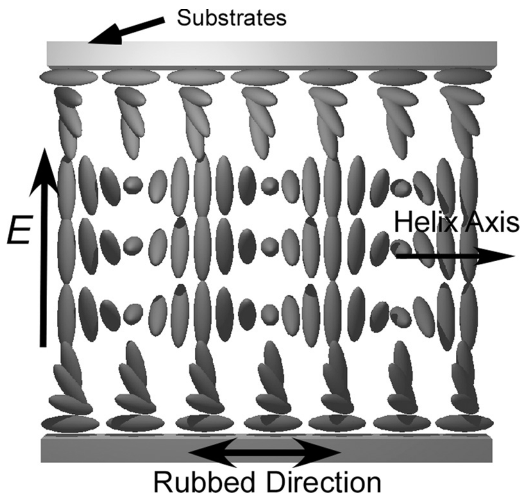

Figure 3. A schematic diagram of the obtained in-plane alignment.

direction, hence forming a helix in the cell-plane direction. A schematic illustration of the obtained in-plane helix alignment is shown in Figure 3.

The emission characteristics of the in-plane helix ChLC were investigated at 29.0°C. Figure 4 shows the emission spectra at the pump energies of 0.2 and 1.6 mJ/cm^2. At low pump energy, spontaneous emission with a broad spectrum was observed. However, a sharp emission peak appeared at 616 nm when the pump energy was 1.6 mJ/cm^2. Figure 5 shows the pumping energy dependence of the emission intensity and the full width at half maximum (FWHM) of the emission peak. The emission intensity linearly increased at a low pump energy, until reaching a threshold pump energy of 0.6 mJ/cm^2, after which the intensity increased dramatically. Concurrently, the broad spectrum was observed with a FWHM of approximately 50 nm was observed below threshold, whereas above threshold a sharp spectral peak appeard with a FWHM narrower than 2 nm (which is the resolutional limit of the spectrometer). These observation indicate that lasing is observed from the dye-doped ChLC with the in-plane helix alignment.

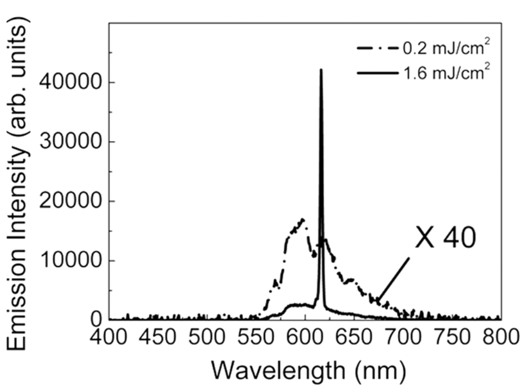

Figure 4. Emission spectra of the in-plane helix alignment at pump energy of 1.6 mJ/cm^2 (solid line) and 200 μJ/cm^2 (dashed line).

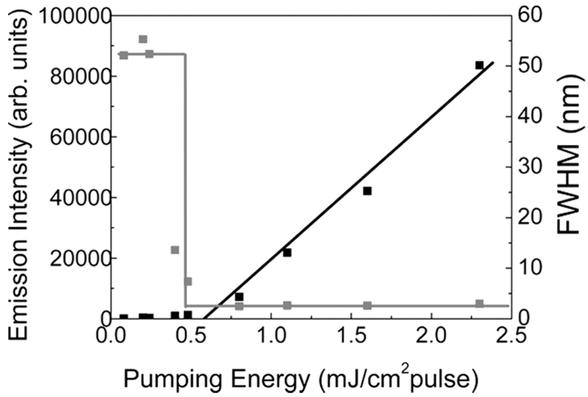

Figure 5. Pumping energy dependence of emission intensity and FWHM.

The electric field dependence of laser action from ChLC with an in-palne helix was investigated at 28.5°C. Figure 6 shows the electric field dependence of the emission spectra. The lasing wavelength shifted toward longer wavelength upon increasing the electric field. The electric field dependence of the lasing wavelength is shown in Figure 7. The lasing wavelength remained almost constant at low fields, but after reaching 3.0 V/μm, it red-shifted continuously with increasing electric field. This can be attributed to the dilation of the helix pitch occurring as a result of applying an electric field perpendicularly to the helical axis [19].

In order to discuss the experimental result, the director distribution of ChLC molecules was calculated upon applying an electric field perpendicularly to the helical axis. We assume that the *x, y*, and *z* components of the LC director is given by $n_x = \cos\theta(z)$, $n_y = \sin\theta(z)$ and $n_z = 0$, where x-axis and z-axis is parallel to the electric field and helical axis respectively. The free energy is given by

$$f = \frac{1}{2}K_{22}\left(\frac{d\theta}{dz} - q_0\right)^2 - \frac{1}{2}\varepsilon_0\Delta\varepsilon E^2\cos^2\theta$$

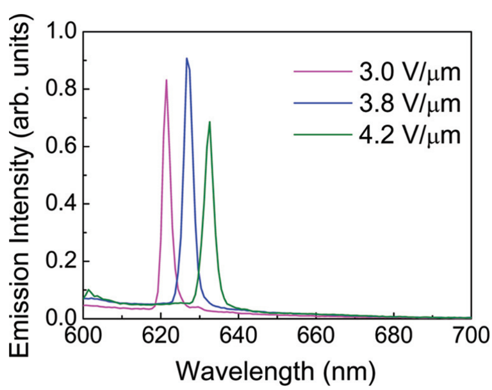

Figure 6. Electric field dependence of emission spectra.

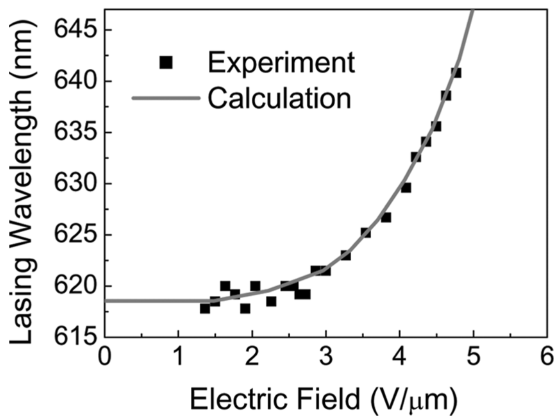

Figure 7. Electric field dependence of measured lasing wavelength (dot) and calculated reflection band edge wavelength (line).

where K_{22} is the twist elastic constant; $P_0 = 2\pi/q_0$, the helical pitch without an applied electric field; $\Delta\varepsilon$, the dielectric anisotropy; and E, the applied electric field [10]. The Euler-Lagrange equation to minimize the free energy is the following,

$$\frac{d\theta}{dz} = q_0 \left[\left(\frac{\pi E}{2E_0} \sin\theta \right)^2 + A \right]^{\frac{1}{2}}$$

where

$$E_0 = \frac{\pi}{2} q_0 \sqrt{\frac{K_{22}}{\varepsilon_0 \Delta\varepsilon}}$$

In Eq. (2) A is the field dependent integration constant, and can be found by solving the equation,

$$\int_0^\pi \left[\left(\frac{\pi E}{2E_0} \sin\theta \right)^2 + A \right]^{\frac{1}{2}} d\theta = \pi$$

From these equations, the electric field dependence of the ChLC director orientation distribution was calculated and is shown in Figure 8. In this calculation, p_0, K_{22}, and $\Delta\varepsilon$ were assumed as 359 nm, 4.8 pN, and 7.5, respectively. While the orientation distribution of the ChLC molecule changed sinusoidally at 0 V/μm, the helical structure deformed and the helical pitch elongated as a stronger electric field was applied. This is understood as a result of the molecules tending to orient along the electric field. The helical structure was unwound completely at the critical electric field of 7.4 V/μm; at this electric field intensity, a dark texture attributed to a homeotropically aligned LC was observed under a POM.

The reflection band edge wavelength was calculated using the pitch length derived from the result in Figure 8. The theoretical fit is plotted in Figure 7, and shows good agreement with the experimentally obtained values. In our experiment, laser action was not observed above the electrical field of 4.8 V/μm, although the

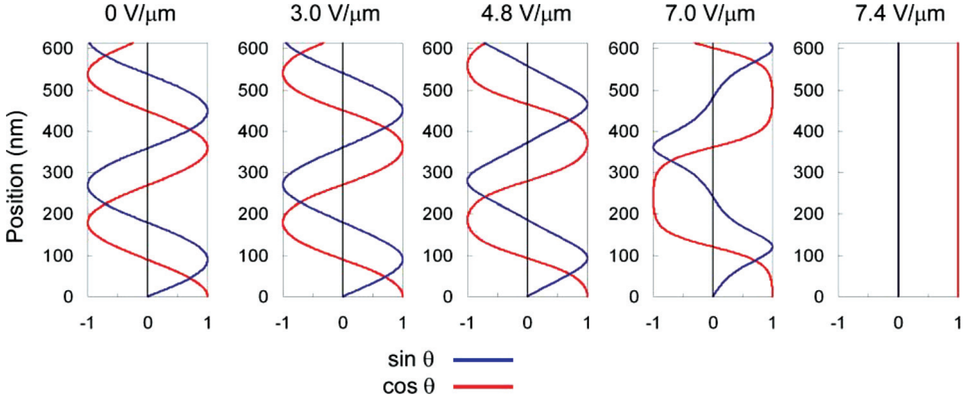

Figure 8. Orientation distribution of CLC molecules as a function of applied electric field. Red and blue line show sin θ and cos θ.

fluorescence spectrum of the DCM dye spans to nearly 700 nm. This is believed to be a result of both weaker fluorescence and the degradation in the cavity quality as the helical structure of the ChLC gradually dilates and deforms. However, we have shown that the helix pitch and hence the lasing wavelength can be tuned continuously by an electric field by using the in-plane helix alignment.

4. Conclusion

ChLCs with an in-plane helix alignment was formed in a conventional rubbed cell by applying an electric field while cooling the sample from the isotropic phase. Laser action in the in-plane helix configuration was observed upon optically pumping the sample and the lasing wavelength could be tuned continuously by varying the electric field intensity. The orientation distribution of the LC molecules in the in-plane helix alignment was calculated at each electric fields and the calculated results of the band-edge wavelengths showed good agreement with the experimentally measured lasing wavelengths.

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