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Position sensitive lasing with continuous wavelength tunability and emission in the cell-plane direction is demonstrated from a photopolymerized cholesteric liquid crystal film. The device has a gradually dilating helix lying in the cell-plane direction and is fabricated by applying a vertical electric field in a conventionally rubbed wedge cell while cooling the sample from the isotropic phase. Tuning range of ~100 nm is achieved by translating the device with respect to the pump beam. Photopolymerizable materials are especially useful in this configuration since a freestanding film, not requiring any external voltage to maintain the molecular ordering, can be prepared. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3089846]

Laser devices based on cholesteric liquid crystals (ChLCs) have been investigated in great detail in recent years in prospect of achieving low-threshold, tunable lasers through a simple self-organization assisted fabrication process.[1] Because of the helical periodic structure, they exhibit selective reflection which is a photonic band gap for circularly polarized light with the same handedness as the material itself.[2] Laser action attributed to Bragg reflection has been observed from the photonic band-edge wavelength, where the group velocity approaches zero and a standing wave forms within the dye-doped medium, leading to an increase in the photon density of states. Mode-hop free single-mode lasing is easily obtained in these systems due to the large separation of usually about 80 nm between the two eigenmodes, i.e., the shorter and longer photonic band-edge wavelength, that are capable of lasing.[3]

Tunable lasing in these systems is due to the sensitivity of LCs to external stimuli, such as heat, electrical field, and light. Since the selective reflection is exhibited over a wavelength range of $n_o p - n_e p$, where n_o, n_e, and p are the ordinary and extraordinary refractive indices of the LC and the helix pitch, respectively, either a change in the helix pitch or the refractive index causes the selective reflectionband and hence the lasing wavelength to shift. However, to obtain a large tuning range, one usually has to rely on the change in the pitch, since only a small change occurs in the refractive index of the material.[4] Tunable lasing has been demonstrated utilizing the temperature dependence of the helix pitch[5] or by doping azobenzene dyes to cause pitch modulation by light irradiation.[6] Lasing at wavelengths spanning over the whole visible range has recently been achieved by forming a spatial pitch gradient by injecting long- and short-pitch ChLCs from two sides of a single cell and photopolymerizing the sample to prevent smearing of the pitch gradient by molecular diffusion.[7] In cases where dynamic tuning of the laser wavelength is not required, creating a pitch gradient inside a single sample and stabilizing the structure seems to be the most efficient tuning method, since tuning is achieved simply by translating the sample with respect to the pump laser beam, without a need to control the temperature or applied voltage.[7–9]

Two of the drawbacks of using a planarly aligned ChLC cell, which is the conventional configuration of the ChLC laser, are that (i) laser emission attributed to the photonic band gap (Bragg mode) is observed only in the cell-normal direction in which the helix axis exists, and quasi-in-plane leaky lasing modes exist in the cell-plane direction due to the waveguide structure formed by the liquid crystal sandwiched between glass substrates, imposing serious limitations on the efficiency of the normal lasing mode,[10] and (ii) only a discrete shift in the pitch and hence the lasing wavelength is allowed because of strong molecular anchoring at the substrate surface.[5] To further enhance the applicability and performance of ChLC lasers, the helix should lie in the cell-plane direction to mitigate the competition between the Bragg and leaky modes, and a continuous, wide-band wavelength tuning is desirable: this is what we demonstrate in this letter. The proposed cell configuration and the conventional planarly aligned ChLC cell are schematically shown in Fig. 1: we fabricate a ChLC laser where the helix lying in the cell-plane direction is gradually dilated within the cell. Laser emission is observed in the cell-plane direction due to the helix lying in the in-plane direction of the cell and continu-

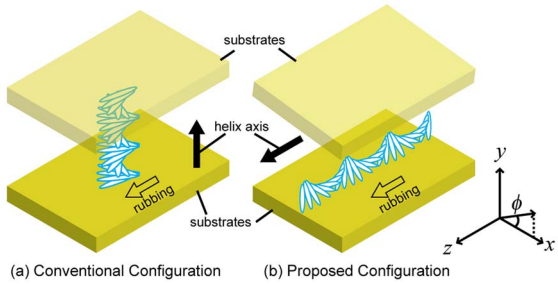

FIG. 1. (Color online) Schematic illustration of the (a) conventional (Grandjean or planar) cell configuration and (b) proposed cell configuration.

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ous wavelength tuning is possible because the helix is not affected by the anchoring of the substrate surface. While there is a previous publication showing that an array of ChLC channels with the helix lying in the cell-plane direction can be fabricated by a holographic lithography process,[11] we show that the in-plane helix alignment of ChLCs can be obtained simply by cooling the sample from the isotropic phase while applying a moderate voltage. We also report tuning of the laser wavelength based on the dilation of the helical structure by electrical fields:[12] a property not demonstrated in the paper previously published.

The sample comprised a photopolymerizable ChLC (RMS03-010) provided by Merck, 1 wt % of photoinitiator irgacure 1850 (Ciba) and 0.6 wt % of laser dye [2-[2-[4- (dimethylamino)phenyl]ethenyl] -6-methyl-4H-pyran-4-ylidene] propanedinitrile (DCM, Exciton). The sample had a right-handed helix and selective reflection band centered at approximately 550 nm at 25 °C. The sample was infiltrated in a planarly rubbed wedge cell with end widths of approximately 3.5 and 7 μm and cooled at 0.5 °C/min from the isotropic phase at 80–25 °C, while applying a rectangular voltage of 30 V (1 kHz). This procedure allowed us to obtain an in-plane helix alignment with the helix axis lying in the direction parallel to the rubbing direction. When an electric field is applied in the direction perpendicular to the helix axis of a ChLC, the director distribution changes to minimize the free energy,

$$f = \tfrac{1}{2}K_{11}(\nabla \cdot \mathbf{n})^2 + \tfrac{1}{2}K_{22}\{\mathbf{n} \cdot (\nabla \times \mathbf{n}) + q_0\}^2 + \tfrac{1}{2}K_{33}|\mathbf{n} \times (\nabla \times \mathbf{n})|^2 - \tfrac{1}{2}\varepsilon_0(\varepsilon_\perp + \Delta\varepsilon \sin^2 \phi)E^2,$$

where $\mathbf{n}$ is the director vector, E is the electric field intensity, $K_{11} \sim K_{33}$ are the splay, twist, and bend elastic constants, respectively, $q_0 = 2\pi/p_0$, where p_0 is the initial pitch length, $\varepsilon_0 = 8.854 \times 10^{-12}$ F/m, $\varepsilon_\perp$ is the ordinary dielectric constant, $\Delta\varepsilon$ is the dielectric anisotropy, and ϕ is the tilt angle of the LC director from the substrate. Assuming the situation described in Fig. 1 where an electric field is applied in the y-direction on a ChLC with its helix lying in the z-direction, and considering a purely twisting situation, the director distribution can be obtained by solving

$$\left(\frac{d\phi}{dz}\right) = q_0\sqrt{A + \left(\frac{\pi}{2}\frac{E}{E_c}\right)^2 \cos^2 \phi},$$

where $E_c = \pi/2 \times q_0 \times (K_{22}/\varepsilon_0/\varepsilon_a)^{0.5}$ is the critical field strength which causes a complete unwinding of the helix and A is a field-dependent constant obtained by solving for[13,14]

$$\int_0^\pi \sqrt{A + \left(\frac{\pi}{2}\frac{E}{E_c}\right)^2 \cos^2 \phi} d\phi = \pi.$$

A particular director distribution is therefore obtained for a given E/E_c value, and as shown in Fig. 2, the helix is gradually unwound and dilated as the applied electric field intensity is increased. The material used in this study had a critical electric field of approximately 8.1 V/μm, which means that E/E_c varied approximately between 1.05 and 0.52. After cell cooling was complete, the sample was irradiated by UV light for approximately 10 s to fix the director configuration by photopolymerization. This procedure allowed us to obtain a freestanding film with a stable in-plane helix configuration even without an applied voltage. Shown in Fig. 3 are the photographs of the photopolymerized sample placed between

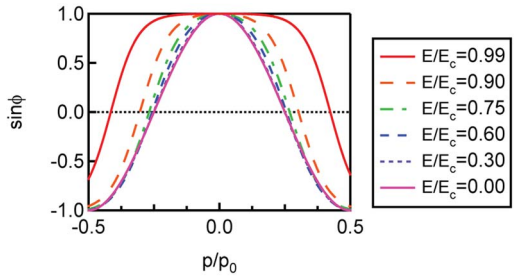

FIG. 2. (Color online) Theoretical deformation of the helical structure of ChLCs calculated at various electrical field strengths with respect to the critical field strength.

crossed polarizers. While no light is transmitted when the rubbing direction is placed at 0° to the crossed polarizers, a gradient of colors are observed when the sample is tilted. This is a characteristic of a short pitched ChLC with an in-plane helix alignment, where the ChLC acts like a uniaxial birefringent material with negative anisotropy with the optical axis in the helix axis direction (see inset Fig. 3). Note that the color change is continuous and no disclination lines are observed due to the anchoring free helix alignment.

Laser measurements were made on the freestanding film placed on a glass substrate. The sample was pumped by a second harmonic from a Q-switched neodymium doped yttrium aluminum garnet laser (Spectron Lasers: SL802) having a wavelength of 532 nm, pulse width of 20 ns, and repetition rate of 10 Hz. The sample was placed on a microscope stage and the pump beam was focused onto a spot of radius of ~10 μm. The sample was translated with respect to the pump beam and the fluorescence was collected in the cell-normal direction by a 20× objective lens (numerical aperture 0.4). Measurements were performed in this configuration for experimental simplicity: we used the well known effect of light being outcoupled to the cell-normal direction via first order Bragg diffraction in distributed feedback lasers.[15] Before investigating the lasing properties, the sample was pumped near the edges and the emission wavelength in the cell-plane and cell-normal directions was confirmed to be equal. A charge coupled device multichannel spectrometer (Hamamatsu Photonics: PMA-11) with a spectral resolution of 2 nm was used to detect the laser emission. To obtain a tunable edge-emitting laser, the direction of the cell thickness gradient should be perpendicular to the rubbing direction: however, the direction of the thickness gradient is essentially unimportant for measurements of the laser emission in the cell-normal direction since the laser is emitted from a finite spot which allows single-mode lasing to be observed even when the helix is dilated in the helix axis

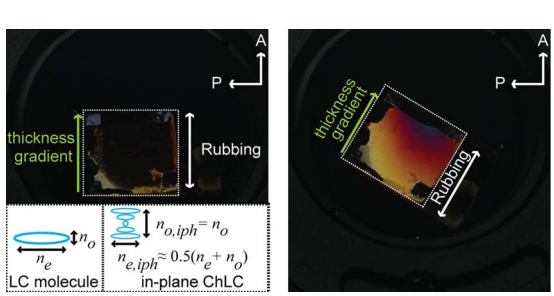

FIG. 3. (Color online) Photograph of the sample placed between crossed polarizers. (Inset) Relationship between the alignment and the observed birefringence.

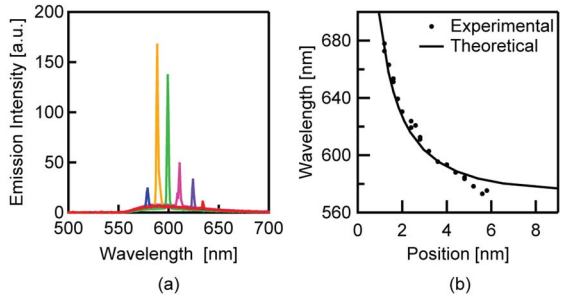

FIG. 4. (Color online) (a) Fluorescence spectra and the (b) lasing wavelengths observed upon pumping the sample at different positions within the sample.

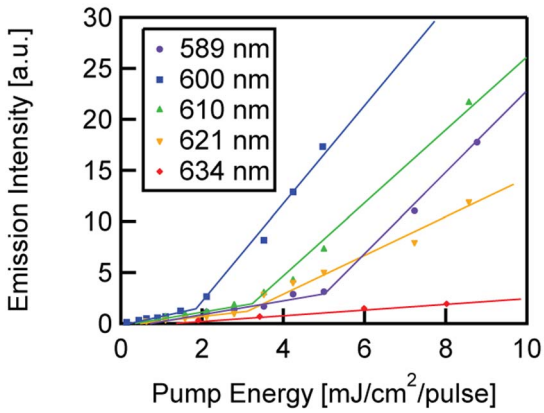

FIG. 5. (Color online) Pump energy dependence of the laser emission intensity at various emission wavelengths.

direction. We fabricated samples with thickness gradients in directions both parallel and perpendicular to the rubbing direction but observed similar wavelength tuning characteristics: here we present results of the sample with the thickness gradient parallel to the rubbing direction.

Figure 4(a) shows the laser emission spectra obtained upon pumping the sample at various positions, with a pump energy of 63 mJ/cm^2/pulse. The lasing wavelengths obtained over a single line along the thickness gradient are shown in Fig. 4(b). As expected from previous discussions, lasing from longer wavelengths were observed from the thinner region of the sample, where the electric field was stronger, and no large stepwise shift of the laser beam is observed as in the case of a conventional planar ChLC. In fact, the emission wavelength could be fine tuned by a resolution of less than 1 nm by measuring the emission spectra at various positions within the cell; we have therefore demonstrated position sensitive continuous wavelength tuning using the proposed cell configuration. The emission wavelength also agreed well with the theoretical prediction [solid line in Fig. 4(b)], which was given by first obtaining the electrical field at a particular position (using the distance from the cell edge), then numerically calculating the deformed helical structure using the experimentally obtained E/E_c value, and finally calculating the transmittance spectrum of the deformed helical structure using the 4×4 formulation[16] with ChLC parameters $n_e=1.72$, $n_o=1.53$, and $p=333$ nm. The tunable range of the laser was about 100 nm, covering approximately the complete fluorescence spectrum of the doped dye. Lasing over a wider range should be attainable by using a combination of dyes, since the helix pitch increases more than twofold at electric field strengths very close to the critical field strength.

Figure 5 shows the lasing threshold characteristics at laser emission wavelengths of 589, 600, 610, 621, and 634 nm. The lasing threshold was lowest at 600 nm, followed by 610, 621, 589, and 634 nm (thresholds are 2.1, 3.5, 3.5, 5.0, and 16.9 mJ/cm^2 pulse, respectively). However, it should be noted that the slope efficiency, given by the constant of proportionality of the emission strength to the pumping energy at regions above the threshold, does not directly correspond to the threshold characteristics. For example, the slope efficiency for 589 nm is higher than for 610 nm, although the lasing threshold is lower at 610 nm. What we have is a very complicated waveguide system, since the efficiencies of the lasing modes are affected by the thickness and the deformation of the helix as well as the gain spectrum of the doped dye. Although we have achieved wavelength tuning, optimization of the film thickness may allow further enhancement of laser performance: numerical modeling of the system using the expressions described in this paper will be discussed in the future.

To conclude, we demonstrated position sensitive tunable lasing from a freestanding ChLC film with an in-plane helix alignment. The proposed configuration is advantageous over the conventional configuration that it allows continuous wavelength tuning and emission of laser light in the cell-plane direction, expanding the applicability of these materials. Using photopolymerizable materials is especially effective in this configuration since a freestanding film can be prepared, not requiring any external voltage once it is fabricated.

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