Reconfigurable Periodic Liquid Crystal Defect Array via Modulation of Electric Field

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AC electric field during the experiment. Corresponding schematic sketches and optical images of the fabricated LC cell are presented in Figure 1a,b. The cell-gap is maintained as 4.5 \( \mu m \). The common LC material, 8CB (4'-n-octyl-4-cyano-biphenyl) is introduced into the LC cell by capillary manner in the isotropic phase. It exhibits a nematic–smectic A phase transition upon cooling from the isotropic phase. The LC director is reoriented by adjusting an AC electrical voltage, \( V = V_0 \sin 2\pi ft \), which leads to generate a uniform LC pattern when the electric voltage reaches a threshold. Here, \( V_0 \) and \( f \) are the amplitude and frequency, respectively. When an AC electric field above \( V_0 = 20 \) V and \( f = 1 \) kHz is applied over the cell, a uniform LC defect array is generated, which are quite different than the typical nematic schlieren texture in the absence of the electric field (Figure 1c). The periodic optical patterns are generated gradually in each unit, identified by a square (yellow dashed line in Figure 1), forming the texture from the electrode edge to the center (yellow arrow) as the voltage is increased (Figure 1d,e; Figure S1, Supporting Information).

The experiment in Figure 1 is carried out under the specific condition of \( V_0 = 30 \) V, \( f = 3 \) kHz, \( T \) (temperature) = 38 \( ^\circ \)C, which corresponds to the nematic phase, using optical microscopy (OM), polarized optical microscopy (POM), and fluorescent confocal polarizing microscopy (FCPM) (Figure S2, Supporting Information). Most of the beautiful optical textures observed
between crossed polarizers show fourfold symmetry and a point defect in the center region (Figure 1d). The detailed LC director distribution can be analyzed by inserting a first order retardation plate ($\lambda = 530$ nm, the slow axis is the blue arrow in Figure 1e), and rotating the sample by 45° (Figure 1e). The black region under the crossed polarizers indicates that the optical axis of the LC molecules, here the director of the 8CB molecule, are oriented vertically to the substrate, or parallel or perpendicular to the polarizers. Blue and yellow colors are observed when the optical axis of the LC is aligned parallel and perpendicular to the slow axis of the retardation plate, respectively, and the LC molecules are arranged concentrically from the each corner to the center (Figure 1d). The Maltese cross patterns are also rotated when the sample is rotated 45° with respect to the crossed polarizers, which confirms the arrangement of the LC molecules (Figure 1e). The point defect is clearly observed by OM as a black dot in the center of the unit (Figure S2a, Supporting Information).

More quantitative analysis could be carried out using FCPM, with a mixture of 0.02 wt%-fluorescent dye molecules $N,N'$-bis(2,5-di-tert-butylphenyl)-3,4,9,10-perylenedicarboximide (BTBP). The bright regions in the FCPM images imply that the LC director is arranged parallel to the linearly polarized excited laser direction (both pink arrows), which changes when the...
excited laser direction rotates, except for the point defect at the center position (Figure 32b, Supporting Information).

Based on these combined results, we recognize that a −1 topological defect resides in the unit of the periodic LC defect pattern.[19–21] The LC director distribution according to the applied electric field is also estimated using commercial software (Techwize LCD 3D, Sanayi System Company, LTD, Korea).[18,22] Each rod represents the average director distribution in top view (Figure 1f) and perspective view (Figure 1g). The vertical orientation of LC director is formed on the electrodes according to the vertically incoming and outgoing electric field. The simulation result indicates that the periodic optical pattern is produced when the LC directors follow the applied electric field, which corresponds to a −1 topological defect in the nematic phase.[19–21] Furthermore, the crossed electrodes regions are considered as +1 topological defect,[21] so that the total topological charge can be conserved as zero. (Figure 1f,g)

2.2. Modulation of the Retardation Color of the LC Pattern

The periodic LC defect array can be modulated by altering the frequency of the applied electric field. The frequency is varied from 1 Hz to 350 kHz at \( V_0 = 30 \, \text{V} \), \( T = 38 \, ^\circ \text{C} \) (Figure 2a). Each LC pattern is stabilized for 5 min before imaging. As the frequency is increased, the retardation color is changed in the respective order of indigo, violet, purple, deep-red, orange, yellow, and white (Figure 2a). This change in interference colors between crossed polarizers coincides with the reduced retardation values in the first order region, which is based on the Michel–Lévy interference chart (Figure 2c).[23] The interference color is achieved as a result of suppressing the light transmission in the specific region of white light. For example, the LC cell shows the purple retardation color by suppressing the light transmission in red colors, as shown at the \( f = 1 \, \text{kHz} \), \( T = 38 \, ^\circ \text{C} \) in Figure 2a,b. Finally, the LC pattern disappears when \( f \) above 300 kHz is used (Figure 2a). This can be explained by dielectric heating, resulting in the phase transition from the nematic phase to the isotropic phase. For \( f > 100\text{kHz} \), a dielectric relaxation process begins, thus the phase shift between the field and the polarization occurs. Then the dielectric heating causes increase in the temperature of the sample, in turn, decreasing the effective birefringence of the LC medium.[24,26] The light retardation \( \tau = 2n_{\text{eff}}d/\lambda \) is proportional to the effective birefringence \( n_\text{eff} \) and thickness \( d \) at a wavelength (\( \lambda \)). In this condition, the cell thickness is fixed as 4.5 µm. Therefore, the light retardation is changed as a function of the effective birefringence of LC, \( n_{\text{eff}} = n_e - n_o \), which is decreased upon heating due to the reduced order parameter, leading to the decrease in the \( n_e \) while \( n_o \) is increased.[21] Here, \( n_e \) and \( n_o \) are extraordinary and ordinary refractive index, respectively. At \( T = 38 \, ^\circ \text{C} \), the \( n_{\text{eff}} \) of 8CB is ~0.13, referring to the previous reports.[27] In this condition, the retardation, which is the multiplication of \( n_{\text{eff}} \) and cell thickness, here ~4.5 µm, is around 590 nm, which exhibits the purple color. As \( f \) is increased, the retardation is decreased because of the reduced \( n_{\text{eff}} \) from ~0.13 to 0. The resulting color sequentially goes through color change from purple to black, which is in good agreement with the reduced average retardation value from 590 to 0 nm.

The identical behavior is observed when the temperature is increased under fixed electric field conditions, \( V_0 \) and \( f \). The color of the LC defect array is changed in the respective order of indigo, violet, purple, deep-red, orange, yellow, and white with increasing temperature from 35 to 40.5 °C under the given electric field condition of \( V_0 = 30 \, \text{V} \), \( f = 3 \, \text{kHz} \) (Figure 2b). Furthermore, the periodic LC defect pattern is faded out at the transition point to the isotropic phase at \( T = 42 \, ^\circ \text{C} \) due to a decrease in \( n_{\text{eff}} \) of 8CB.[27] Accordingly, the color change triggered by a temperature seems to be similar to the color variation induced by changing the applied frequency (Figure 2a,c). The \( n_{\text{eff}} \) value is decreased from 0.14 at \( T = 35 \, ^\circ \text{C} \) to 0 at \( T = 41 \, ^\circ \text{C} \), which is consistent with the dielectric heating effect observed in the frequency varying experiment (Figure 2a).

2.3. Optical Vortex Beam Generation

A well-controlled LC defect array fabricated over a large area can serve as a good optical vortex inducer based on the concept of the Pancharatnam–Berry (PB) geometric phase.[28] At a certain condition of temperature and electric field, the demonstrated LC cell has the optical path difference of the ordinary and extraordinary waves to act as a half-wave plate for a beam with a wavelength of 632.8 nm. A half-wave plate inverts the handedness of the circularly or elliptically polarized light and the rotation of the fast axis of the half-wave plate by an angle \( \Theta \), providing the transmitted light with a relative phase shift of 2\( \Theta \). The arrangement of half-wave plate elements as shown in Figures 1 and 2 delivers a helical distribution of the relative phase of the transmitted circularly polarized light; when a right-handed (or a left-handed) circularly polarized (RCP (or LCP)) beam passes through the LC cell, the Laguerre–Gaussian beam is generated with a left-handed (or a right-handed) circular polarization (LCP (or RCP)) and a topological charge of \( +2 \) (−2).[29] To analyze the geometrically controlled phase distribution of the transmitted vortex beam, the interference pattern with a reference beam is examined (Figure 3a). Here, the reference beam should have the same circular polarization to the transmitted Laguerre–Gaussian beam through the LC defect. The laser beam is illuminated on the unit cell with a beam diameter of 40 µm in the full-width at half-maximum (Figure 3b). The beam profiles and interference patterns of the output beams are observed at \( V_0 = 30 \, \text{V} \) with varying frequency and temperature (Figure 3c–g). The intensity profile at \( f = 3 \, \text{kHz} \), \( T = 38 \, ^\circ \text{C} \) exhibits a donut shape, which is a typical vortex beam profile having a central phase singularity (Figure 3c). The topological charge of the vortex beam can be estimated from the interference patterns. The fork-shaped interference pattern with two dislocations confirms that the LC cell generates the optical vortex with a phase topological charge of \( +2 \) (−2) for RCP (or LCP) incident beam, which confirms that the resultant LC cell is a q-plate with a topological charge of −1 (Figure 3c).[30] In addition, the actuation of the vortex beam could be switched off by either increasing frequency (Figure 3d) or temperature (Figure 3e) due to the phase transition to the isotropic phase. Interestingly, the intensity profile of the vortex beam is weakened and distorted with decreasing temperature because of the increased retardation of incident light, while the LC structure...
is maintained (Figure 3f). Further reduction in the temperature induces a highly distorted intensity profile, as a result of the transition to the smectic A phase whose short range order no longer allows for formation of a vortex beam (Figure 3g).

The optical path difference between extraordinary and ordinary waves, which depends on LC configuration, LC film thickness, and $\Delta n_{\text{eff}}$, determines the vortex beam generation in transmission. As mentioned above, the highest efficiency of the conversion to the vortex beam is acquired when the LC component acts as a half-wave plate for the used wavelength. In our case using a 632.8 nm wavelength laser, the most uniform and high-intensity vortex beam can be generated when the LC cell induces about 300 nm retardation to the light, which exhibits the first order retardation color in yellow by suppressing the
light transmission in near-ultra violet regions. In the meantime, the first order yellow retardation color in the LC pattern appears at the isotropic-nematic transition point. Thus, the highest efficient beam conversion is found at this temperature. In our 4.5 µm LC sample, the most uniform and high-intensity vortex beam is formed at T = 40 °C and f = 3 kHz. (Figure 3; Figure S3, Supporting Information). Since Δn_{eff} of the LC molecule is easily changed depending on the f or T, we expect that the reconfigurable LC defect can also act as a wavelength-tunable q-plate.

2.4. Particle Trapping and Releasing Actuation

To show the versatility of our work, the fabricated LC defects are used to trap and release small particles in the defect core, by controlling the free energy.[6,7,31,32] In our experiments, a LC-particle composition made of 8CB and 2 µm sized silica spheres is introduced in the LC cell. Figure 4 shows the time sequence of trapping (Figure 4a) and releasing (Figure 4b) of the silica particle in an LC defect core.

When the LC defect is generated, embedding a −1 topological defect, the randomly distributed particle gradually moves toward the point defect (Figure 4a; Video S1, Supporting Information). Thus, when the LC orientation is arranged to form the −1 defect patterns in response to the applied electric field, the microparticles are elastically trapped into specific site, here the defect cores where the distortion of LC orientation is the largest.[32] This defect-mediated localization of the microparticles assures that the overall free energy cost is minimized.[32] Furthermore, the −1 defect structure is erasable when high frequency or high temperature is used as shown in Figure 2. Here, we apply f = 500 kHz in order to avoid the spontaneous reorientation of LC molecules near the isotropic-nematic transition. This implies that modulation of the LC pattern would also induce the release of the particle from the defect structure (Figure 4b; Video S2, Supporting Information). It is caused by the transient effect related to relaxation of the defect structure upon turning LC pattern off.[32] The assembly of the particle, therefore, is affected by the presence and absence of defect structure, which can be accomplished by varying the experimental conditions. The reversibility of the reconfigurable LC defect is directly measured using a 2D CCD camera (see Videos S1 and S2 in the Supporting Information).

2.5. Versatile Topological LC Defect Patterns

More complex topological defects can be created when different sets of electrodes are combined to form a unit cell (Figure 5).[17] A schematic sketch and optical image of the electrode configuration are presented in Figure 5a,b. The blue and red indicate the top and bottom electrodes, which are used to apply the AC electric field. The light-blue and violet lines indicate the interdigitated electrodes, where the electric field is not applied (Figure 5a). And the cell-dimensions are slightly changed so that the unit cell had a width of 10 µm and space between electrodes of 10 µm. Figure 5c shows a POM image of distinct structures generated from the diverse combination of electrodes, achieved in a single cell, at V_0 = 30 V, f = 3 kHz, T = 38 °C. The detailed textures are also found in two small boxes at the bottom in each cell, showing the molecular arrangement after inserting the full wave plate and rotating the sample 45°. In addition, the LC distribution and the optical simulation results show the equivalent LC orientation as those predicted by the POM experiment (Figure S4, Supporting Information).

As described so far, the Maltese-cross optical pattern with fourfold symmetry, which shows a −1 topological defect at
the center point, appears in the crossed linear electrodes (ii): top-right part in Figures 1, 2, and 5c). The LC molecules are oriented vertically in the intersecting part of the interdigitated electrodes, and are distorted along the electrodes, due to electrostatic induction, although the electric field is not applied to these light-blue and violet electrodes (iii–(iv) in Figure 5c).

An asymmetrically distorted molecular arrangement is generated in the single interdigitated electrode, forming a two-fold symmetry-LC pattern (ii) and (iv) in Figure 5c). The LC pattern maintains a structure of the point defect and −1 winding number, but the fourfold symmetry is broken by the hybrid electric field induced by the single interdigitated electrode (iii).

Figure 5. Various periodic and uniform LC patterns achieved by alternating combinations of crossed electrodes under an electric field at $V_0 = 30$ V and 3 kHz, 38 °C. a) Schematic diagram and (b) OM image of a LC sandwich cell assembled to have a crossed electrodes configuration. The AC electric field is applied from the blue (top) and red (bottom) electrodes. c) POM images of the various periodic LC defect structures (left inset: with the first order retardation plated inserted; right inset: under 45° rotated sample.
and (iv) in Figure 5c and Figure S4 (Supporting Information)). When two interdigitated electrodes are added, the vertical alignment of LC is induced at the crossed electrodes, and vertical and horizontal orientations are simultaneously induced to form a fourfold symmetric configuration in a unit cell (iii) in Figure 5c and Figure S4 (Supporting Information). Therefore, the high pre-tilt orientation is maintained, and the birefringence is reduced compared to other LC patterns (i), (ii), and (iv) in Figure 5c. In addition, while maintaining –1 geometrical configuration, the point defect disappears due to the vertical orientation of the intersection of the electrodes at the center and the escaped configuration (iii) in Figure 5c and Figure S4 (Supporting Information)). Thus, these structural and optical LC defect arrays including the defect shapes can be fabricated and controlled by varying configuration of electrodes.

Interestingly, the optical textures our LC cell can be further manipulated by varying cell thickness, in which the retardation value, its tunable range, and the curvature of molecular arrangement can be changed as a result of the level of vertical confinement (Figure S5, Supporting Information). Noted by this result, our system is possible to broaden the range of wavelength-tunable plates and even convert the Laguerre–Gaussian beam profile to azimuthal mode as well as radial mode.

3. Conclusion

In summary, we fabricated reconfigurable periodic LC defect arrays using crossed electrodes. This system enables the formation of various LC defect patterns on a single cell without any additional surface treatment and expensive optical processes. The topological defects generated here have –1 strength and are modulated by changing either the frequency of the applied field or temperature. The individual LC defects can be employed to demonstrate an optical vortex inducer and a particle trapping-releasing unit, which could be dynamically controlled. Furthermore, various types of periodic LC arrays could be generated by adjusting the combination of electrodes. The proposed platform can be easily and widely applied in lithographic applications as well as optoelectronic devices. Indeed, this electric field-driven optical structure generation is not limited in the specific LC phase, here the nematic phase, but applicable to many other LC phases including cholesteric LC and twist bend nematic LC phases.

4. Experimental Section

Materials and Sample Preparation: LC cells were prepared using patterned ITO substrates with striped electrodes of 10 µm width and 30 µm spacing. LC cells having 2.5, 4.5, and 9.5 µm thicknesses were obtained using silica beads as spacers, and the exact thickness of the cells was measured by Feby–Perot interference method. The cell was filled with a commercial LC material, 8CB (4′-n-octyl-4-cyano-biphenyl, Synthon Chemicals) via capillary force at its isotropic temperature (T > 40.5 °C). The temperature of the sandwich cells was controlled with a heating stage (LINKAM LTS420, Tadworth, UK) equipped with a temperature controller (LINKAM TMS94, Tadworth, UK). For FCPM observations, the 8CB LC was mixed with 0.02 wt% of a fluorescent dye molecule, BTBP (Aldrich). To demonstrate the particle trapping, 1 wt% of 2 µm silica particle was mixed with 8CB, which is filled into the LC cell.

Imaging and Simulation of LC Structure in LC Sandwich Cell: The optical textures of the LC cells were observed by POM (LV100POL, Nikon) with a CCD camera (DS-R1i, Nikon). The FCPM (C2 plus, Nikon) investigation was performed with a linearly polarized laser source (λ = 488 nm, Coherent). The LC director distribution with a top-down applied AC electric field was calculated using commercial software (Techwiz LCD 3D, Sanayi System Company, LTD., Korea).[18]

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

This work was supported by a grant from the National Research Foundation (NRF) and was funded by the Korean Government (MSIT) (2017M3C1A3013923, 2018R1A5A1025208, and 2017R1E1A1A01072798).

Conflict of Interest

The authors declare no conflict of interest.

Keywords

defect array, electric field, liquid crystal, modulation, reconfigurable

Received: May 29, 2019
Revised: July 24, 2019
Published online: September 18, 2019

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