Unusual double four-lobe textures generated by the motion of carbon nanotubes in a nematic liquid crystal

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Abstract: Unusual double four-lobe nematic liquid crystal (LC) textures were observed in the carbon nanotube (CNT)-doped nematic LC under electric field. Through the electro-optical studies in a wide range of vertical electric fields in the direction of the long axis of the LC molecules, it was realized that the double four-lobe nematic LC textures were formed in the range of 120 to 160 V_{rms} at 1 Hz. The formation of these unusual double four-lobe nematic LC textures could originate from the electric field-induced movement of CNTs and the subsequently frustrated reorientation of LCs.

OCIS codes: (160.3710) Liquid crystals;(250.4110) Modulators

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1. Introduction

Liquid crystal (LC) devices are ubiquitous because of their principal attractions such as low power consumption, high resolution, thinness, lightness, and flatness. The performance of a LC device strongly depends on the LC materials used. Therefore, numerous efforts have been attempted to change the essential electro-optical properties of the existing LCs by introducing nano-particles [1-3]. Among these nano-particles, the nanoscaled carbon materials have become a topic of major research due to their outstanding electrical properties and strong interactions with aromatic mesogenic units of LCs [4-6]. In fact, under a low voltage, the electro-optical properties of a twisted nematic cell and an in-plane switching LC cell were significantly improved by introducing a small amount of CNTs [7-14]. While in the high voltage regime, different LC textures were observed possibly due to field-induced motions of CNT in the nematic LC medium [5, 6, 15]. Various textures of nematic LC droplets embedded in polymer matrix induced by periodic distortions have also been reported [16, 17].

In this paper, we report on the formation of unusual double four-lobe nematic LC textures in the CNT-doped LC under an ac electric field. Based on the electro-optical experiments in a wide range of vertical electric field, it was concluded that the double four-lobe nematic LC textures could originate from the electric field-induced movement of CNTs and the subsequently frustrated reorientation of LCs. In addition to the insight into the fundamental physics of CNT-doped nematic LC, this study provides new opportunities to LC engineers to design and fabricate novel LC or photonic devices.

2. Experimental

The nematic LC mixture (MJ951160) employed in this study was purchased from Merck and used as received. Its physical properties are listed as follows: dielectric anisotropy $\Delta\varepsilon = 7.4$, birefringence $\Delta n = 0.088$ at the incident light wavelength $\lambda = 589$ nm, and nematic phase between -40 and 87℃. The LC cell consists of two glass substrates of which inner surfaces are coated with a thin and transparent indium-tin-oxide (ITO) electrode. To achieve a vertical alignment, a thin polyimide layer (AL-60101 from Japan Synthetic Rubber Co.) was coated on the ITO-glass substrates.

The thin multi-walled CNT (t-MWCNT) was synthesized by the catalytic chemical vapor deposition using FeMoMgO catalyst. The t-MWCNT had number of tube walls of 2 ~ 6 with the corresponding diameters of 3 ~ 6 nm, an inner diameter of 0.7 ~ 2.6 nm [18]. The t-

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MWCNT sample was shortened by chemical cutting using H_2SO_4/HNO_3. Through filtering with deionized water (pore size = 0.45 μm) and centrifuging in dichloroethane (DCE) solvent (at 8000 rpm for 30 min), the processed t-MWCNT had an average length of 250 nm [4].

The t-MWCNT in a powder type was not dispersed instantly in LC medium. In order to disperse t-MWCNT in LC medium, 10^{-3} wt% of t-MWCNT was dissolved into DCE solvent, followed by mixing with nematic LC. If the proper conditions such as sonicating time and power were not satisfied, t-MWCNTs would aggregate themselves during the dispersion process. After the solvent evaporation, we measured the clearing point of LC mixture to confirm the solvent evaporated perfectly. This gave rise to the same value as the pure LC. The t-MWCNT-dispersed LC mixture was thoroughly sonicated for an hour and filled into the LC cell by capillary action at room temperature. Without further treatments, the vertical LC alignment was achieved in the cell with a 60 μm gap and the CNT clusters were not observed at least in a microscopic level.

Polarized optical microscope (POM, Nikon L-UEPI, Japan) with a digital camera (Nikon DMX1200, Japan) was utilized to observe the LC texture evolutions which depend on the applied electric voltage of the sinusoidal waves.

3. LC textures under vertical electric fields

We have previously reported the dynamic behavior of CNT-doped nematic LCs under a vertically aligned LC cell [6, 15]. Above a critical electric field, CNTs can perturb the LC alignment due to their enhanced translational motions, resulting in light leakage from the LC cell. In fact, the directors of the LC with a positive dielectric anisotropy should align parallel to the vertically applied electric field, resulting in a perfect dark state under crossed polarizers. Nevertheless, the light leakage associated with deformation of the LC director is generated by the translational motion of CNTs. Based on the density functional calculations, we realized that the charge is transferred from LC to CNT [19]. When the CNT-doped LC is placed in an ac electric field, the large dipole moment is induced in the CNTs due to their high aspect ratio and CNTs experience the dielectrophoresis (DEP) force [15]. In case of a permanent dipole moment of CNTs due to an excess charge transfer from LC to CNTs, similar ideas can be adopted as follows. Considering one dimensional translation motion of CNTs in nematic LC medium and applied electric field as a sinusoidal wave, the velocity (v) and displacement (x) of CNTs in LC medium at a time can be written as [15]:

$$\mathrm{v} = (E_0 - E_{cr})\sqrt{\frac{2\Gamma\varepsilon_m \operatorname{Re}\{K(\varepsilon^*_{cnt}, \varepsilon^*_m)\}}{m}}\sin(\omega t) \qquad (1)$$

$$x = \frac{2(E_0 - E_{cr})}{\omega}\sqrt{\frac{2\Gamma\varepsilon_m \operatorname{Re}\{K(\varepsilon^*_{cnt}, \varepsilon^*_m)\}}{m}}\left(\sin^2\frac{\omega t}{2}\right) \qquad (2)$$

where Γ is a geometrical factor [15], ε^*_{cnt} and ε^*_m are complex dielectric permittivities of the nanotube and LC medium, respectively. $K(\varepsilon^*_{cnt}, \varepsilon^*_m)$ represents the complex polarization factor (for spherical objects this is known as Claussius-Mossotti function). E_0 denotes the applied ac field and E_{cr} is the critical electric field at which the CNTs start motion. For ωt is an odd multiple of π, the displacement of the CNTs in LC medium will be maximum (x_{max}). Therefore amplitude (*Ampl*) i.e. maximum displacement of the translation motion of the CNTs in LC medium can be written as

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$$Ampl = \frac{2(E_0 - E_{cr})}{\omega}\sqrt{\frac{2\Gamma\varepsilon_m \operatorname{Re}\{K(\varepsilon^*_{cnt}, \varepsilon^*_m)\}}{m}} \tag{3}$$

On the other hand, in the vertically aligned LC layers under crossed polarizers, the light leakage in the cell can be described as follows [5]:

$$T / T_o = \sin^2 2\psi(V,t)\sin^2(\Gamma_{eff}(V,t)/2) \tag{4}$$

where ψ is the angle between the LC director and the polarizer's transmission axis and Γ_{eff} is the effective cell retardation. In Eq. (4), ψ and Γ_{eff} are functions of both voltage and time, which implies that the LC textures could change according to the applied voltage and time evolution.

When a relatively high sine-wave voltage (60 Hz) was longitudinally applied to the CNT-doped nematic LC cell, the light leakage was observed as four-lobe nematic LC textures when viewed through POM, as shown in Fig. 1(A). These four-lobe nematic LC textures in the POM were observed only above the threshold voltage of 60 V_{rms}. The number of four-lobe nematic LC textures was increased with the strength of electric field with different sizes. This phenomenon could originate from the fact that the dispersed CNTs in the nematic LC medium are not monodisperse in length, therefore the mass (*m*) and *q/m* ratio (here, *q* is the net excessive charge of the CNT) is not the same. As a result, the critical electric field needed for the CNT motion and the subsequent light leakage is dependent on the *q/m* ratio of the CNTs, since the acceleration of CNTs is proportional to *E q/m* [5]. All the observed four-lobe nematic LC textures disappeared when the applied electric field was removed, as shown in Fig. 1(B).

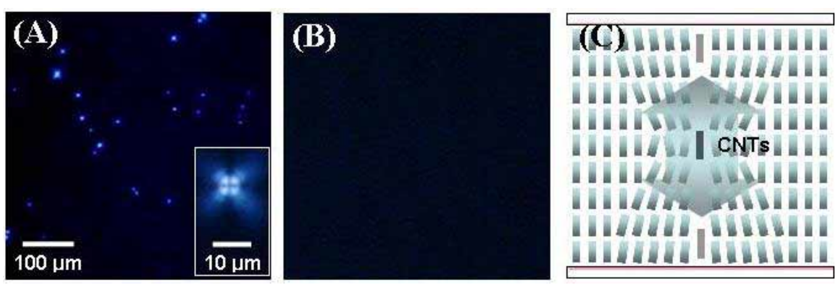

Fig. 1. POM microphotographs of a t-MWCNT-doped longitudinally aligned LC cell between crossed polarizers at (A) V = 150 V_{rms} with 60 Hz, and (B) V = 0, which was released right after applying 150 V_{rms}. (C) is the deformed LC directors. The inset in Fig. 1(A) shows the magnified image of a four lobe texture.

If the applied voltage is higher than the threshold voltage (at a frequency of f = 60 Hz), due to the fast up and down motion of CNTs, the deformed LCs shown in Fig. 1(C) may have sufficient time to reorient themselves in tune with the CNTs motion. Hence the shape of the four lobe texture once formed seems not to be changed throughout the cycle of the applied sinusoidal wave voltage because of limited time resolution of human eyes. However, if the applied voltage is in the range of 120-160 V_{rms} with f = 1 Hz, a peculiar double four-lobe texture was observed.

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The voltage dependent LC texture change was systematically investigated by taking snap shots of POM images. 15 pictures were taken per second and the results are displayed in Fig. 2. The frequency of the sinusoidal electric field was kept at 1 Hz. At V~60 V_{rms}, no distinct patterns were observed. On the other hand, at V>160 V_{rms} four-lobe nematic LC textures were formed at regular intervals of time and phase angle of the sine wave electric field input. Surprisingly, at the intermediate electric fields between 120 and 160 V_{rms}, unusual double four-lobe nematic LC textures were observed, which was emphasized as a broken red line in Fig. 2.

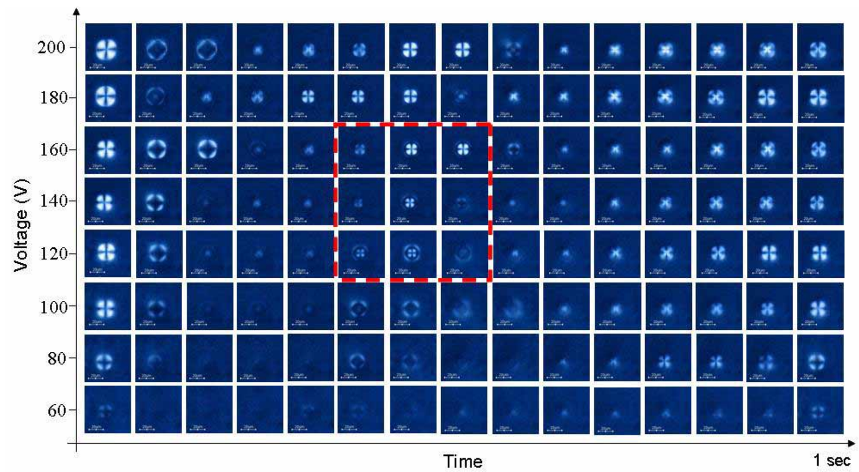

Fig. 2. POM microphotographs of four-lobe textures generated in a vertically aligned nematic LC cell with increased ac voltage (with f = 1 Hz). The region within the broken red line shows the double four-lobe nematic LC textures.

According to the equations (1) and (3), at low electric field (below 60 V_{rms}), the CNTs start to translate with a low velocity and amplitude so that little deformation of LC molecules occurs. This leads to light leakage in the form of indistinct textures due to small Γ_{eff} in a local area, as shown in Fig. 2. However, above the upper critical electric voltage (160 V_{rms}), CNTs move with fast (see Eq. (1)) up and down between the electrodes and deform the LC molecular arrangements, resulting more light leakage in a form of clear four-lobe nematic LC textures due to relatively large Γ_{eff} in a wider area than that in 60 V_{rms}. According to the Eq. (3) the amplitude of the translational motion of the CNTs will also be large at high electric field, i.e., CNTs translate from the bottom electrode to the top electrode during the positive electric field cycle. The deformation of the LC directors is then symmetrically generated along the moving direction at both sides, as shown in Fig. 3(A). On the other hand, during the negative electric field cycle, the CNTs move back to the original position, deforming the LC orientation in a reverse way, as shown in Fig. 3(B).

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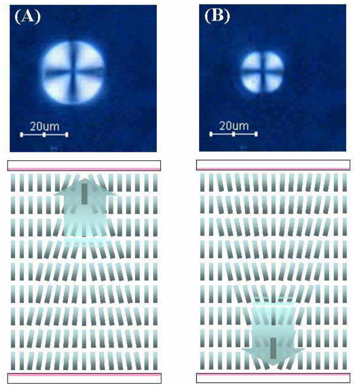

Fig. 3. POM microphotographs and LC orientations of the CNT-doped nematic LC cells at 180 V_{rms} with 1 Hz: (A) upward movement of CNT and (B) downward movement of CNT.

When the applied voltage is between 120 and 160 V_{rms}, then from Eq. (3) the amplitude of translational motion corresponding to this voltage range would be smaller than that at 160 V. Therefore, in one cycle of the applied electric field, the CNTs travel from the bottom electrode and return back somewhere in the mid way before reaching the top electrode. During the positive half cycle, the CNTs translate towards the high electric field and the pressure wave in LCs is generated due to the motion of CNTs. Before this developed pressure wave disappears, the CNTs generate another pressure wave in an opposite direction induced by reversing the applied electric field direction and the superimposition of two pressure waves takes place. This results in the twin LC molecular orientations in both upper and lower sides of the CNTs, as illustrated in Fig. 4(B).

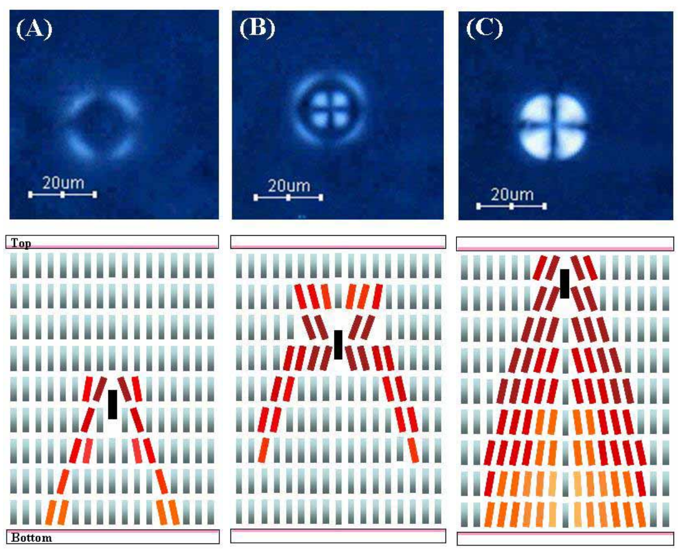

Fig. 4. (2.46 MB) Movies of POM and LC orientations of the CNT-doped LC cells in the middle of a cycle at (A) 80 V_{rms}, (B) 120 V_{rms}, and (C) 200 V_{rms} with 1 Hz sine wave frequency. Assuming that a CNT starts to move from the bottom electrode by applying electric fields, the deformed LC directors are represented by reddish colors.

As illustrated in Fig. 4(A), the LC molecules are not deformed much, since the the velocity and amplitude of CNTs are low under the relatively low electric voltage such as 60~100 V_{rms}. On the other hand, CNTs in the nematic LC medium translate so fast at 200 V_{rms} that the nematic LC texture appears clearly due to larger deformations of the LC molecules than that

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in 80 V_{rms}, as shown in Fig. 4(C). The anomalous twin orientation of LC molecules at the intermediate electric fields between 120 and 160 V_{rms} could be the origin of the formation of unusual double four-lobe nematic LC texturesDuring the single cycle at 140 V_{rms} with f = 1 Hz, the more detailed processes of CNT translations in the nematic LC medium under the electric fields are illustrated in Fig. 5 with LC director deformation pictures (lower parts) and their corresponding light leakage patterns (upper parts). As mentioned earlier at this voltage, the CNTs translate somewhere in the mid way before reaching the top of the electrode. According to Eq. (2), the maximum displacement of the CNTs' motion will correspond to a phase angle of 180° of the input voltage due to a factor of $\omega t/2$ i.e., at this phase angle the velocity of the CNTs would be zero (see Eq. (1)). However, the displacement would be maximum. Now consider the case when the phase angle of the input applied field is 90°. At this phase angle, the velocity would be maximum and the displacement would be half the maximum. Therefore, the perturbation of CNTs on LC and hence the induced pressure on LCs would be maximum and as a consequence there will be the maximum light leakage. Fig. 5(A) shows the maximum light leakage in the form of four lobe texture. Therefore at this position the CNTs is somewhere at the distance near to half the maximum with their maximum velocity. The light leakage is diminished with further increase in the phase angle of the input wave because of decreasing velocity and resulting low perturbation force at LC molecules up to the phase angle of 180° (see figs. 5(B) and (C)). Before this developed pressure wave disappears, due to change in the polarity in the next half cycle of the applied electric field, CNTs translate in the reverse direction, and hence generate another pressure wave in the opposite direction and both pressure waves superimpose. Thus the frustrated double four-lobe texture is generated (see Fig. 5(D)). With further increase in the phase angle and time, the previously developed pressure wave disappears and the velocity of CNTs and hence perturbation of CNTs on LCs again increase in the reverse direction. Therefore the light leakage in the form of four lobe texture again starts growing (see Figs. 5(E) and (F)). Perminov et al. [20] reported transient motion and aggregates of nanoparticles due to dipole-dipole interaction using electromagnetic light wave. In our case of CNTs at 1 Hz, we have also observed the similar translational motion due to the DEP forces [21] rather than aggregate effect with dipole-dipole interaction in high frequency region.

Finally, the LC textures are also observed as a function of the frequency of an applied voltage and the texture observed did not show the double four-lobe textures at the same time resolution. With increasing frequency the amplitude of the translation motion of CNTs will decrease as indicated in eq. (3) and the polarity of applied voltage changes rapidly resulting in less possibility of existence of similar frustrated reorientation because of fast translational motion of the CNTs. Consequently, the same type of unusual double-lobe texture (at 1 Hz) associated with frustrated reorientation was not clearly observed at the same time resolution. On the other hand, the persistence of the measured equipment was 15 frames/sec and the director of the LCs is distorted more than the recognizing efficiency of above equipment. Hence double four-lobe texture was not clearly observed at high frequency especially greater than 15 Hz.

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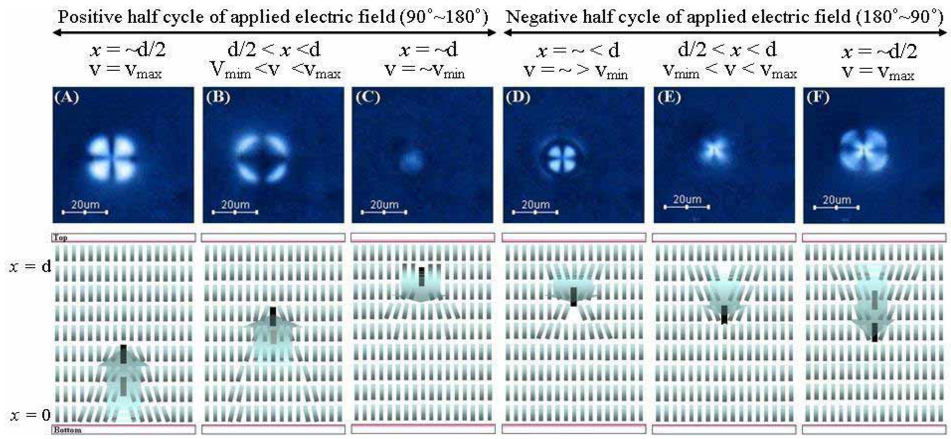

Fig. 5. Process of the LC director reorientations during the single cycle at 140 V_{rms} and f = 1 Hz. The d represents the amplitude of translation motion of CNTs in LC medium and d/2 is the distance corresponds to the half of the amplitude of translation motion of CNTs in LC medium for 140 V_{rms} and f = 1 Hz.

4. Conclusion

The light leakage in the CNT-doped longitudinally aligned nematic LC cell under various applied ac voltages was investigated. Below the lower critical electric voltage 60 V_{rms} with 1 Hz sine waves, no distinct patterns were observed while four-lobe nematic LC textures were formed above the upper critical electric voltage (160 V_{rms}). Surprisingly, at the intermediate electric fields between 120 and 160 V_{rms}, the unusual double four-lobe nematic LC texture was observed. Based on the systematic electro-optical experiments in a wide range of applied electric fields and their LC director calculations, we conclude that the unusual double four-lobe nematic LC textures are formed due to the deformation of the LC molecular alignments in two different directions at a particular instant and phase angle. In addition to the insight into the fundamental physics of CNT-doped nematic LC, this study about the movements of CNTs under the applied ac electric fields can pave new pathways for the manipulation of CNTs for nano-device applications.

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