Electro-Optical Bistability Associated With Stripe Formation in Highly Twisted Nematic Liquid Crystals
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An electro-optical (EO) bistability associated with the stripe formation is studied in highly twisted nematic liquid crystals (NLCs) as a function of the ratio d/P0 of the cell thickness d to the natural pitch P0 in the presence of an applied voltage. For d/P0 ≥ 0.6, it is found that the EO hysteresis appears in the π-twisted planar geometry. Particularly, for d/P0 ≥ 0.75, a transition from the π-twisted state to the 2π-twisted state occurs together with the EO hysteresis. The origin of the EO bistability comes from the in-plane modulation of the molecular reorientation which leads to the formation of the stripe domains. The experimental data of the stripe period and the threshold for the appearance of the stripe domains are analyzed in terms of the periodicity of the in-plane modulation within the continuum theory. [DOI: 10.1143/JJAP.43.5435]

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1. Introduction

The electro-optical (EO) properties of twisted nematic liquid crystals (NLCs) result mainly from the distortions of the helicoidal structure by the application of an external electric field. Because of the intrinsic periodicity presented in the helicoidal structure, the nature of instabilities, phase transitions, and the associated relaxation processes in highly twisted NLCs are quite complex.1,2) A variety of transitions to the modulated states having in-plane stripes or ripples have been experimentally observed3–8) and theoretically analyzed9,10) Moreover, a theoretical approach to a multi-critical behavior of the ripple instability was presented in a cholesteric LC.11) A twisted NLC confined in a finite region, for example, a twisted NLC cell made with parallel substrates, is of great importance to fabricate NLC-based optical devices and information displays. For the NLCs with positive dielectric anisotropy, when an external electric field is applied to the direction of the helix of the twisted NLC, the molecules rotate toward the field direction and thus the helix will be distorted.12) The resultant EO properties depend on the natural pitch and the optical anisotropy. In fact, the twisted structure itself is governed by the ratio d/P0 of the cell thickness d to the natural pitch P0 of the twisted NLC.7,8) As a consequence, for highly twisted NLCs such as cholesteric LCs, it is expected that the EO hysteresis occurs due to the generation of a certain type of a modulated state. For instance, in cholesteric LCs, Chigrinov et al. investigated the problem of the threshold appearance of spatially periodic deformations under the action of an external electric field.2) Hurault et al. calculated the period of the modulated structure from the free energy density of a deformed cholesteric LC.13)

In this paper, we studied the EO bistability associated with the generation of the modulated structures in highly twisted NLCs as a function of d/P0 in the presence of an applied voltage. The observed EO bistability together with the EO hysteresis is found to originate from the stripe formation depending on the magnitude of the initial twist. The EO characteristics of the modulated structures including the stripe period and the threshold behavior have been measured as a function of the applied voltage for different values of d/P0. In the presence of an external electric field, the existence of the bistability was optically verified by the formation of stripes in twisted NLCs previously.14,15) Our experimental data of the period and the threshold for the modulated structure as a function of d/P0 agree well with theoretical results.

2. Experimental

The NLC material used in this work was the ZLI-2293 mixture of Merck, doped with the chiral agent of S-811 to produce a highly twisted structure. The dielectric anisotropy and the elastic constants of the ZLI-2293 mixture are Δε = 10.0, K1 = 12.5 × 10−12 N, K2 = 7.3 × 10−12 N, and K3 = 17.9 × 10−12 N, respectively.16) The natural pitch of the twisted NLC can be determined by the weight concentration c of the chiral dopant from P0 = (cHTP)−1 where P0 is a natural pitch of the twisted LC and HTP is a helical twisting power. Here, HTP is given by about 10 μm.

Highly twisted NLC cells were made up of conductive indium-tin-oxide glasses coated with polyimide of AL1051 (Japanese Synthetic Rubber Co.) for planar alignment with low pretilt angle (±2°). The internal surfaces of the cell were unidirectionally rubbed so as to give the π-twisted orientation of the NLC molecules. The thickness of each cell was maintained using the glass spacers of 7 μm. Each assembled cell was placed between crossed polarizers such that the rubbing direction made the angle of 45° with respect to one of crossed polarizers. The EO transmission through the highly twisted NLC cell was measured under a square-waveform voltage at the frequency of 100 Hz. For each twisted NLC cell, the value of P0 was determined by the Cano-wedge cell method.12) A He-Ne laser of 632.8 nm, an arbitrary waveform generator, and a digital multimeter were used for measuring the EO properties of the twisted LC cells. The applied voltage was first increased to 10 V and then decreased to 0 V for the EO hysteresis measurements.

Microscopic textures of the cells were observed using a polarizing optical microscope under crossed polarizers in the presence of the applied voltage. The stripe domains were

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formed above a certain value of $d/P_0$ at about the Fredericks transition. The periodicity and the direction of the strips were determined for various values of $d/P_0$ on increasing and decreasing the applied voltage. All the measurements were carried out at room temperature.

3. Results and Discussion

In a twisted planar geometry, the twist angle of the LC molecules with respect to one of the rubbing directions depends on the ratio $d/P_0$. When the molecules on two rubbed substrates are aligned along the rubbed directions at both surfaces, the $\pi$-twisted configuration is stable if the ratio $d/P_0$ satisfies $\Phi_T/\pi < d/P_0 < 2\pi$, where $\Phi_T$ is a twist angle of the NLC.

In our $\pi$-twisted cell, the transmitted light intensity was measured as a function of the applied voltage for different values of $d/P_0$ on increasing and decreasing the applied voltage. The ratio $d/P_0$ was varied from 0.29 to 1.0 to produce one of two highly twisted configurations with $\Phi_T = \pi$ or $\Phi_T = 2\pi$. Figures 1(a) and 1(b) show the transmitted intensities for $d/P_0 = 0.59$ and 0.62, respectively. For $d/P_0 = 0.59$, no hysteresis exists and the transmission curve is single-valued as shown in Fig. 1(a). For $d/P_0 = 0.62$, however, the EO hysteresis appears between a certain voltage range, marked by two gray arrows, on increasing and decreasing the applied voltage. From the fact that the EO hysteresis exists, it is expected that a transition to an optical bistable state appears above a certain threshold in a highly twisted configuration.

In the absence of an applied voltage, the LC molecules are aligned parallel to the substrates and twisted continuously from one substrate to another because of the elastic distortions. When the applied voltage above the Fredericks transition is on, the LC molecules with positive dielectric anisotropy tend to reorient perpendicular the cell surfaces and the twisted structure becomes unwound. When the voltage is off, the LC molecules normally become relaxed into the initial state with no hysteresis as shown in Fig. 1(a). However, if two different distorted states corresponding to the voltage-up and the voltage-down processes are involved during relaxation into the initial state, a hysteresis would appear like in our case as shown in Fig. 1(b). Note that the existence of such EO hysteresis is directly governed by the magnitude of the ratio $d/P_0$. In Fig. 2, we present experimental results for the voltage range where the EO hysteresis exists as a function of $d/P_0$. It is clear that in highly twisted planar geometry with $d/P_0 > 0.6$, the EO hysteresis appears. One interesting point is that the maximum voltage range for the hysteresis was observed for $d/P_0 \approx 0.75$.

We now describe the physical origin of the EO hysteresis observed in the highly twisted planar geometry with $d/P_0 \gtrsim 0.6$. In a less twisted geometry with $d/P_0 < 0.6$, the voltage-down process will follow reversibly the path of the voltage-up process during relaxation into the initial state and thus no hysteresis exists. In a more twisted geometry with $d/P_0 \gtrsim 0.6$, however, the distorted state is relaxed into the initial state through a certain intermediate state created by the high degree of twist. This is quite similar to the appearance of the periodic textures in a cholesteric LC. In the cholesteric LCs aligned uniformly, the in-plane structure becomes modulated, and stripe or finger print domain appears in the texture. This means that the formation of in-plane modulated states such as the stripe domains in our case produces the EO hysteresis in the highly twisted geometry with $d/P_0 \gtrsim 0.6$ since splay distortions are not energetically favorable.

As discussed above, in the highly twisted regime of $d/P_0 \gtrsim 0.6$, the maximum EO hysteresis was observed at $d/P_0 = 0.75$ where a transition from the $\pi$ twisted state to the $2\pi$ twisted state occurs.

Figures 3 and 4 show microscopic textures of the highly twisted LC cells with $d/P_0 = 0.62$ and 0.77 under an applied voltage of 2.5 V, respectively. For $d/P_0 = 0.62$, the stripe
domains shown in Figs. 3(a) and 3(b) were obtained on increasing and decreasing an applied voltage. In both cases, the direction of the stripes is conserved and parallel to the rubbing direction. The white arrows (M) represent the direction of the LC molecules in the midplane of the cell. The period of the stripes in the voltage-down state is larger than that in the voltage-up state.

In a more highly twisted LC with $d/P_0 > 0.75$, the direction of the stripes on increasing the voltage makes a right angle with respect to that on decreasing the voltage as shown in Figs. 4(a) and 4(b). Figure 4 shows microscopic textures of the highly twisted NLC cell with $d/P_0 = 0.77$ at the applied voltage of 2.5 V. Note that the maximum EO hysteresis associated with the in-plane modulated structure in the highly twisted geometry was observed at $d/P_0 > 0.75$ where a transition for the twist, changing the direction of the stripes, occurs. In the $\pi$-twisted state ($d/P_0 < 0.75$), the LC molecules are aligned perpendicular to the rubbing direction in the midplane of the cell due to the continuous deformations of the molecules. As the voltage increases, the splay deformation is induced in the middle of the cell and the LC molecules are periodically modulated along the LC director because of the boundary conditions. The direction of stripes is parallel to the rubbing direction but perpendicular to the molecular director since the stripes have the same phase modulation.

For $d/P_0 > 0.75$, on the other hand, the $2\pi$-twisted state is energetically more stable since the twisting power in the highly twisted NLC overcomes the elastic energy produced by the distortions of the molecules bounded on the unidirectionally rubbed substrates. In fact, the surface pretilt plays a critical role in the competition of the helical twisting power with the boundary conditions. For a higher pretilt, the $2\pi$-twisted state can not appear because of the excess elastic energy but a transition for the twist is possible for a lower pretilt angle of about $5^\circ$. Hence, in the $2\pi$-twisted state, the LC director in the middle of the cell is parallel to the rubbing direction and the direction of stripes is mostly perpendicular to the rubbing direction as shown in Fig. 4(a). As the voltage decreases, however, the direction of stripes is parallel to the
rubbing direction, like in the $\pi$-twisted state of the highly twisted NLC, as shown in Fig. 4(b). This is similar to a transition for the twist observed in a bistable twisted NLC cell.\(^{18}\) It may be noted that the $\pi$- and splayed $2\pi$-twisted states are not topologically equivalent and there exist an energy barrier due to the presence of disclinations. In this case, the splayed $2\pi$-twisted state can not be transformed continuously into the vertical state and the transition between two states needs an activation energy. In the high field regime, the electric field energy works as an activation energy and the LC molecules rotate toward the vertical state which is topologically equivalent to the $\pi$-twisted state.

Let us describe the modulated, stripe structure in the highly twisted LC using the two dimensional distribution of the NLC director, $\mathbf{n}$. The free energy density of the twisted NLC is given by,\(^{12}\)

$$F = \frac{1}{2}(\nabla \cdot \mathbf{n})^2 + \frac{K_2}{2}(\mathbf{n} \cdot \nabla \times \mathbf{n} + q_0)^2$$

$$+ \frac{K_3}{2}(\mathbf{n} \times \nabla \times \mathbf{n})^2 - \frac{1}{2}D \cdot E,$$  \hspace{1cm} (3.1)

where $K_1$, $K_2$, and $K_3$ are the splay, twist, and bend elastic constants, respectively. The wave vector is represented by $q_0 = 2\pi/P_0$ and $D$ and $E$ are the displacement and electric field vectors. Suppose that the cell thickness is expressed in terms of $z$ and the in-plane modulation occurs along the $x$-axis. The LC director at the point of $(x, z)$ is described in spherical coordinates by the polar angle $\theta$ and the azimuthal angle $\phi$. In principle, the LC director distribution in the equilibrium state is obtained by minimizing the free energy. In the presence of an external electric field $E$ along the $z$-axis, $\theta(x, z)$ and $\phi(x, z)$ can be expanded in the harmonic series of $x$ and $z$ provided that the electric field is not too strong. For a relatively weak field, we assume that the azimuthal angle $\phi(x, z)$ remains constant and ignore the higher order terms of the polar angle $\theta(x, z)$. The polar angle is then written as

$$\theta(x, z) = \theta_0 \sin \left( \frac{\pi x}{w} \right) \sin \left( \frac{\pi z}{d} \right),$$  \hspace{1cm} (3.2)

where $w$ is a spatial period of the in-plane modulation and $\theta_0$ is the maximum polar angle which is identical to the tilt angle in the midplane of the cell. Substituting eq. (3.2) into the expression of the free energy density eq. (3.1) and minimizing the resultant free energy with respect to the parameter $w$, the period of the in-plane modulation is obtained by

$$w^2 = \frac{1}{4} P_0 d \left( \frac{3K_3}{2K_2} \right)^2.$$  \hspace{1cm} (3.3)

Figure 5 shows the period $w$ of the stripes as a function of the ratio $d/P_0$. Open circles and the solid line represent the experimental data and the numerical results obtained from eqs. (3.3), respectively. For numerical simulations, $K_2 = 7.3 \times 10^{-12}$ N and $K_3 = 17.9 \times 10^{-12}$ N were used. It is clear that the numerical results agree well with the experimental data.

Under the condition that the elastic energy is comparable to the electric field energy, the threshold voltage for the in-plane modulation, i.e., the formation of the stripes, appears is given by\(^{23}\)

$$V_{th}^2 = \frac{8\pi^3}{\Delta \epsilon} \left( \sqrt{6K_2K_3} + 4K_3d \left( \frac{1}{p} - \frac{1}{P_0} \right) \right) \frac{d}{P_0},$$  \hspace{1cm} (3.4)

where $\Delta \epsilon$ is the dielectric anisotropy and $p$ is the actual pitch of the twisted NLC in the cell. Note that in a confined structure, the pitch $p$ is generally different from the natural pitch $P_0$. Figure 6 shows the threshold voltage for the appearance of the stripe domains in the highly twisted NLC cell as a function of $d/P_0$. Open circles and solid lines represent the experimental data and the numerical results obtained from eq. (3.4), respectively. In the calculation, for the ZLI-2293, the literature values of the elastic constants and the dielectric anisotropy were used. There exists a discontinuity in the threshold voltage at $d/P_0 = 0.75$ which corresponds to the transition from the $\pi$-twisted state to the $2\pi$-twisted transition. Again, this is consistent with the EO hysteresis behavior as a function of $d/P_0$ as shown in Fig. 2. The $2\pi$-twisted state requires more electric energy to produce the in-plane modulation because of the higher elastic deformations of the LC molecules. Hence, the periodicity of the in-plane modulation decreases with
increasing $d/P_0$ and the twisted state becomes more unstable in the presence of an applied voltage. This tells us that for the highly twisted NLC in a confined geometry, the deformation of the helix itself in addition to the unwinding of the helix plays an important role in the EO bistability.

4. Concluding Remarks

In the highly twisted NLCs, the EO bistability associated with the in-plane modulation in a form of stripes were observed for relatively large values of $d/P_0$ on increasing and decreasing an applied voltage above the Fredericks threshold. The period of the stripes decreases with increasing $d/P_0$. The maximum EO hysteresis was found at $d/P_0 = 0.75$ where a discontinuity in the threshold voltage for the stripe formation exists. The discontinuity corresponds to a transition from the $\pi$-twisted state to the $2\pi$-twisted state. Our experimental data of the stripe period and the threshold voltage for the appearance of the in-plane modulation are in excellent agreement with the numerical results obtained in the continuum theory. The work presented here would benefit understanding the essential features of the in-plane modulation in highly twisted NLCs and optimizing twisted NLC-based devices such as super-twisted NLC displays.

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