Electro-Optical Hysteresis by the In-plane Modulation of the Molecular Reorientation in Highly Twisted Nematic Liquid Crystal Devices

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Abstract

We studied on an electro-optical (EO) hysteresis, coming from the in-plane modulation of the molecular reorientation, in highly twisted nematic liquid crystal devices. Varying a ratio d/P_0 of the cell thickness d to the natural pitch P_0, the EO hysteresis and the twisted angle transition were observed and analyzed in terms of the periodicity of the in-plane modulation within the continuum theory.

1. Introduction

Twisted nematic liquid crystals (NLCs) confined in a finite region are of great importance to fabricate NLC-based optical devices and information displays. The electro-optical (EO) properties of the twisted NLCs are mainly governed by the distortions of the helicoidal structure by an applied external field. In highly twisted NLCs, a variety of transitions to the modulated states having in-plane stripes and ripples in the present of an applied external field have been experimentally observed [1-6] and theoretically analyzed [7,8]. Moreover, a theoretical approach to a multicritical behavior of the ripple instability was presented in a cholesteric LC [9].

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 [10]. 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/P_0 of the cell thickness d to the natural pitch P_0 of the twisted NLC [5,6]. 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 [11]. Hurault *et al.* calculated the period of the modulated structure from the free energy density of a deformed cholesteric LC [12].

In this paper, we studied the EO hysteresis associated with the generation of the modulated structures in highly twisted NLCs as a function of d/P_0 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/P_0. Our experimental data of the period and the threshold for the modulated structure as a function of d/P_0 agree well with theoretical results.

2. Experiments

Highly twisted NLC cells used in this work were made up of conductive indium-tin-oxide glasses coated with polyimide of AL1051 (Japan 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. The NLC material 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 $\Delta\varepsilon = 10.0$, $K_1 = 12.5 \times 10^{-12}$ N, $K_2 = 7.3 \times 10^{-12}$ N, and $K_3 = 17.9 \times 10^{-12}$ N, respectively [13]. The natural pitch P_0 of the twisted NLC can be determined by the weight concentration c of the chiral dopant from $P_0 = (c\ HTP)^{-1}$ where HTP is a helical twisting power. Here, HTP is given by about 10 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 P_0 was determined by the Cano-wedge cell method [10]. 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. 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 π-twisted configuration is stable if the ratio d/P_0 satisfies $\Phi_T/2\pi - 0.25 < d/P_0 < \Phi_T/2\pi + 0.25$ where Φ_T is a twist angle of the NLC. In our π-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. Figs. 1 (a) and (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 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 $d/P_0 \approx 0.75$.

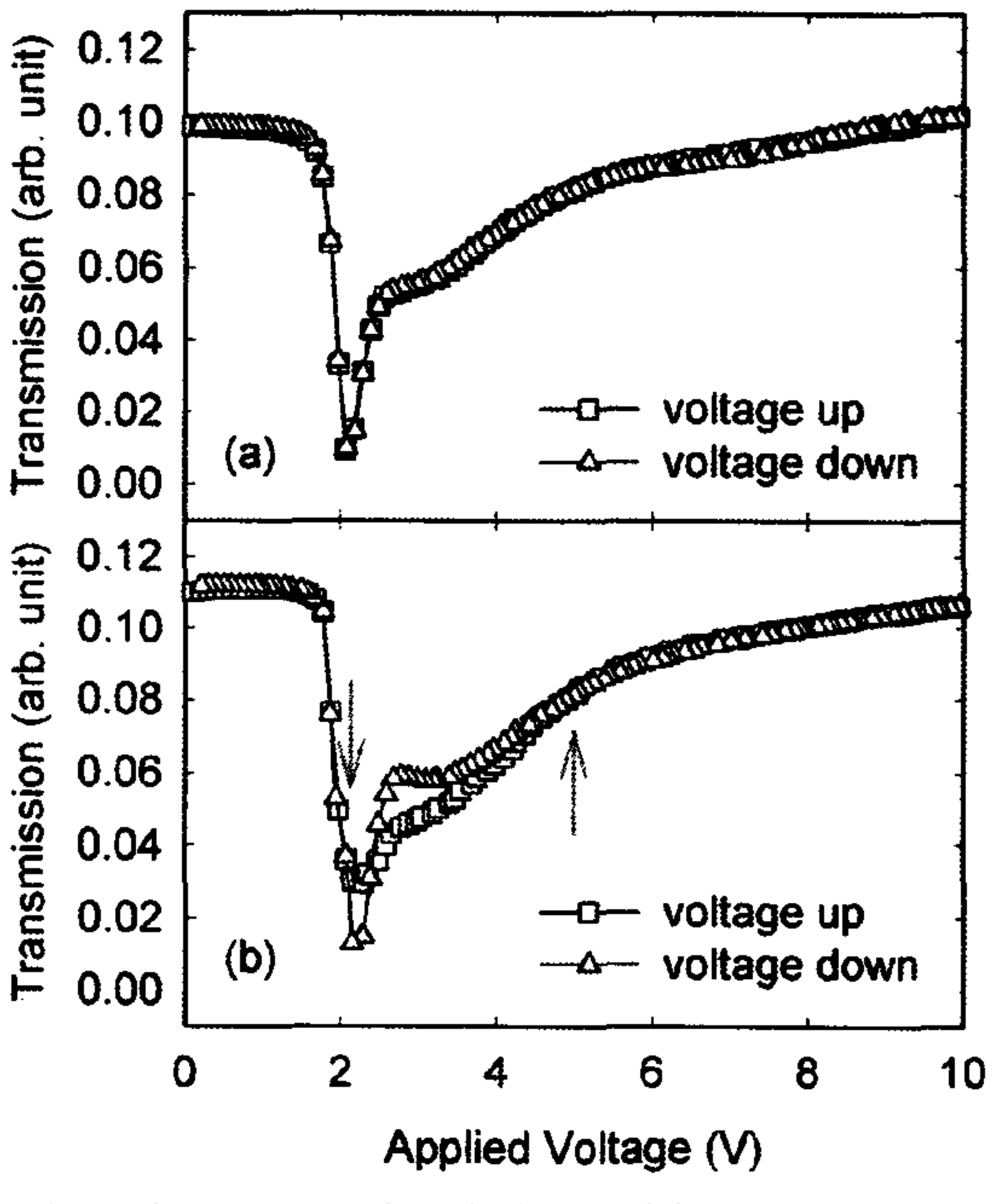

Fig. 1. The transmitted intensities of the highly twisted NLC cells with the ratio d/P_0 of (a) 0.59 and (b) 0.62 as a function of the applied voltage. Gray arrows indicate the voltage range of the EO hysteresis on increasing and decreasing the applied voltage.

We now describe the physical origin of the EO hysteresis observed in the highly twisted planar geometry with $d/P_0 \geq 0.6$. 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 $d/P_0 \geq 0.6$, 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 [14]. 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 $d/P_0 \geq 0.6$ [5] since splay distortions are not energetically favorable [9]. In the highly twisted regime of $d/P_0 \geq 0.6$, the maximum EO hysteresis was observed at d/P_0

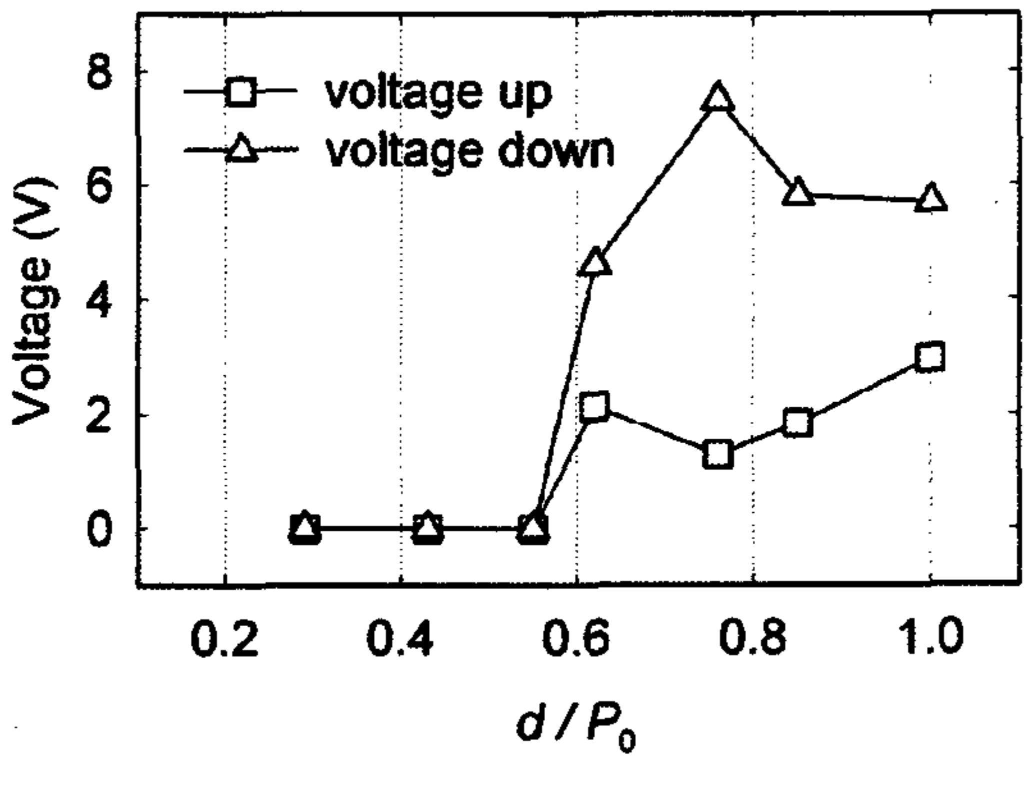

Fig. 2. The characteristic voltages for the appearance of the EO hysteresis as a function of d/P_0. Open rectangles and triangles correspond to the voltage-up state and the voltage-down state on increasing and decreasing the applied voltage, respectively.

= 0.75 where a transition from the π-twisted state to the 2π-twisted state occurs.

Figure 3 shows 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 Fig. 3(a) and (b) were obtained on increasing and decreasing the voltage, respectively. The twisted NLCs become modulated in the plane parallel to the substrates above a certain threshold. The stripe direction on both increasing and decreasing the voltage is parallel to the rubbing direction but the period of the stripe on increasing the voltage is larger than that on decreasing the voltage. It should be emphasized that as shown in Figs. 3(a) and (b), the stripes are always extended along the rubbing direction and independent of the voltage-on and voltage-down processes for d/P_0 = 0.62.

In a more highly twisted LC with $d/P_0 \geq 0.75$, the direction of the stripes on increasing the voltage makes a right angle with respect to that on decreasing the voltage. Figs. 3(c) and (d) show 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 $d/P_0 \geq 0.75$ where a transition for the twist, changing the direction of the stripes, occurs. For $d/P_0 > 0.75$, the 2π-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 [15]. In the 2π-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. 3(c). As the voltage decreases, however, the direction of stripes is parallel to the rubbing direction, like in the π-twisted state of the highly twisted NLC, as shown in Fig. 3(d). This is similar to a transition for the twist observed in a bistable twisted NLC cell [16].

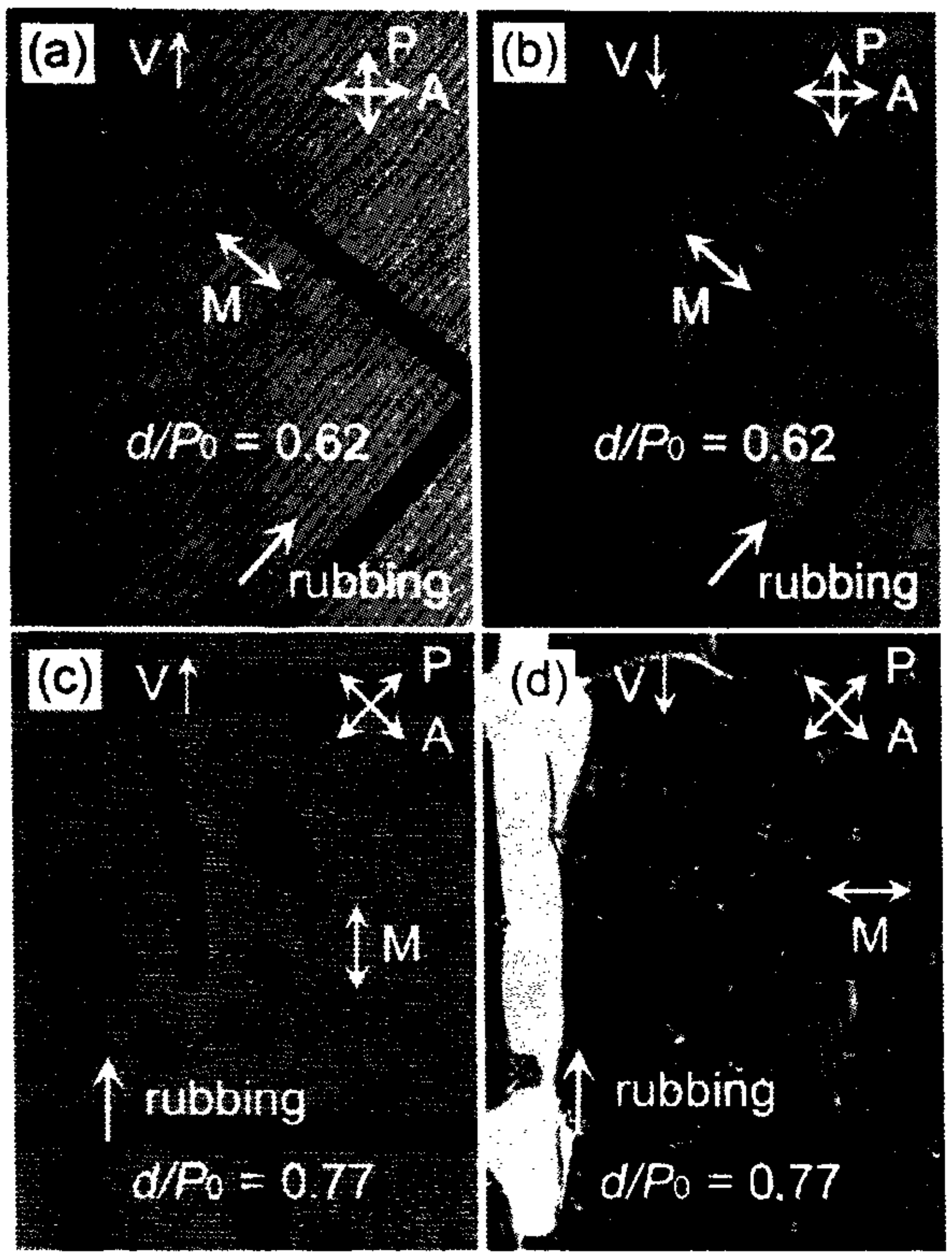

Fig. 3. The stripe textures of the highly twisted NLC cells with d/P_0 = 0.62 [(a) and (b)] and 0.77 [(c) and (d)] observed at 2.5 V under crossed polarizers: (a) and (c) increasing, (b) and (d) decreasing an applied voltage, respectively. The white arrows (M) represent the direction of the LC molecules in the midplane of the cell.

Let us describe the modulated, stripe structure in the highly twisted LC using the two dimensional distribution of the NLC director. 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 θ and the azimuthal angle ϕ. 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 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 written as

$$\theta(x,z)=\theta_m \sin\left(\frac{\pi z}{d}\right)\sin\left(\frac{\pi x}{w}\right), \qquad (1)$$

where w is a spatial period of the in-plane modulation and θ_m is the maximum polar angle which is identical to the tilt angle in the midplane of the cell. Substituting Eq. (1) into the expression of the free energy density and minimizing the resultant free energy with respect to the parameter w, the period of the in-plane modulation is obtained by

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

Moreover, 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, appear is given by [11]

$$V_{th}^2 = \frac{8\pi^3}{\Delta\varepsilon}\left[\sqrt{6K_2K_3} + 4K_2d\left(\frac{1}{p} - \frac{1}{P_0}\right)\right]\frac{d}{p}, \qquad (3)$$

where $\Delta\varepsilon$ 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.

Figures 4(a) and (b) show the period w of the stripes and the threshold voltage for the appearance of the stripe domains as a function of the ratio d/P_0. Open symbols and the solid line represent the experimental data and the numerical results obtained from Eqs. (2) and (3), respectively. In the calculation, for the ZLI-2293, the literature values of the elastic constants and the dielectric anisotropy were used. It is clear that the numerical results agree well with the experimental data. In Fig. 4(b), there exists a discontinuity in the threshold voltage at d/P_0 = 0.75 which corresponds to the transition from the π-twisted state to the 2π-twisted transition.

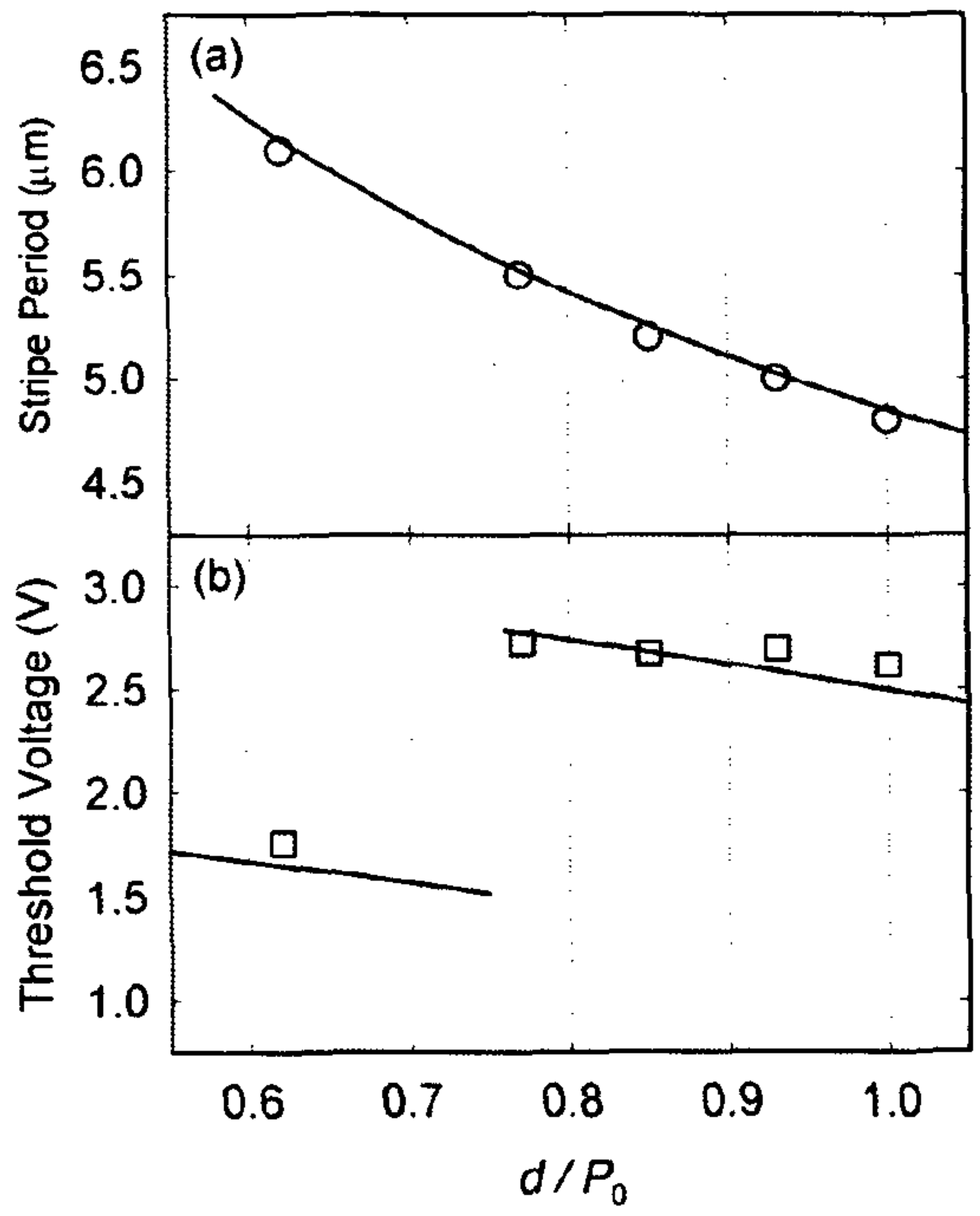

Fig. 4. (a) The stripe width (period) and (b) the threshold voltage for the appearance of the stripes as a function of d/P_0. Open symbols and the solid line represent the experimental data and the numerical results, respectively.

4. Conclusion

In the highly twisted NLCs, the EO hysteresis associated with the in-plane modulation were observed for relatively large values of d/P_0 on increasing and decreasing an applied voltage. 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 π-twisted state to the 2π-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 NLC displays.

5. Acknowledgement

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