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Electrohydrodynamic Instability in Cholesteric Liquid Crystals in the Presence of a Magnetic Field

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*We report electrohydrodynamic instability in a cholesteric liquid crystal—a mixture of *p*-methoxy benzylidene *p*-*n*-butylaniline (MBBA) and a cholesteric dopant cholesteryl nonanoate (CN), in the presence of a superimposed magnetic field H . We have found a certain intensity H^* with respect to the instability in this mixture with negative dielectric anisotropy, beyond which a stripe pattern appears. With an increasing electric field, the stripe pattern evolves into a grid pattern (GP). Investigations of the H -dependences of thresholds and wavelengths for these patterns in comparison with those of nematic liquid crystals clearly show that the typical GPs found in cholesteric liquid crystals are a superposition of two kinds of stripe patterns induced by the electrohydrodynamic instability.*

Keywords: cholesteric liquid crystal; electrohydrodynamic instability; grid pattern; pitch

INTRODUCTION

Ac-driven electrohydrodynamic instability that occurs in nematic liquid crystals (NLCs) has been extensively investigated both theoretically and experimentally [1–2]. This instability provides us with a rich variety of stationary and nonstationary patterns. The most typical pattern, the Williams domain (WD), is induced by the so-called Carr-Helfrich effect [3]. Increasing the strength of the electric field and changing its frequency produces various secondary instabilities such as the zigzag instability and abnormal roll instability [4–5]. Various

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other patterns occur as well, which depend on the initial alignments of the director \mathbf{n} [6], where \mathbf{n} denotes a unit vector defining a locally averaged direction of the elongated molecules of liquid crystals.

This study investigates cholesteric liquid crystals (CLCs) with negative dielectric anisotropy [7–11]. Chiral molecules have an additional order along the helical axis \mathbf{h} , which is always perpendicular to \mathbf{n} . Introducing a helical pitch p characterizes CLCs. In principle, CLCs ($p \neq \infty$) can be considered as a type of NLC ($p = \infty$) with a finite p . In CLCs, however, the pitch serves as a critical factor in understanding its electrohydrodynamic instability and resulting pattern formation. Typical to CLCs is a gridlike pattern (GP); however, CLCs exhibit a variety of patterns depending on p [10–11].

This study investigates the threshold characteristics and wavelength of GPs in CLCs with a well-controlled p . In particular, it examines the change of patterns and the variation of their thresholds in the presence of a superimposed magnetic field, comparing the results to those of NLCs. These experimental results may help in understanding the pattern-formation mechanism and the role of pitch p in CLCs. In particular, the results clearly explain the formation of the typical GPs found in CLCs.

EXPERIMENTAL RESULTS AND DISCUSSION

This investigation used a cholesteric liquid crystal (CLC) made from mixing *p*-methoxy-benzylidene *p*-*n*-butylaniline (MBBA) with a small amount of a cholesteric dopant, cholesteryl nonanoate (CN). We prepared several MBBA-CN mixtures with 0.1 wt.% to 5.0 wt.% of CN. In this concentration (c) range, the CLC's pitch (p) varied from 120 to 2.4 μm . The actual pitch was determined using the relation $pc = 0.12 \pm 0.01 \mu\text{m}$ ($0 < c \leq 0.05$) [9]. The so-called helical twisting power ($1/pc = \text{const.}$) is important for this study as well as other applications of CLCs [16–17].

The experiments were performed on sandwich-type cells whose glass surfaces were coated with transparent electrodes (indium tin oxide). The electric conductivities and dielectric constants for the MBBA used (at 25°C) were $\sigma_{\parallel} = 6.50 \times 10^{-8} \Omega^{-1} \text{m}^{-1}$, $\sigma_{\perp} = 3.11 \times 10^{-8} \Omega^{-1} \text{m}^{-1}$, $\varepsilon_{\parallel} = 4.67$, and $\varepsilon_{\perp} = 5.32$, respectively. The gap distance between the electrode surfaces, which are treated for the planar alignment of the director, was $d = 50 \mu\text{m}$, and the lateral (active) cell size was $1 \times 1 \text{ cm}^2$.

Across these cells, an alternating electric field $\mathbf{E} = E_0 \cos(2\pi ft)$ [$\mathbf{E} = (0, 0, \pm E_z)$] was applied. The field was generated by function synthesis (NF-1915) and amplified using an Amplifier (F20A, Toyo Tech), as shown in Figure 1(c). A magnetic field, \mathbf{H} , controlled by an electromagnet system (EMD-6, EMIC) was also applied parallel to the glass plates

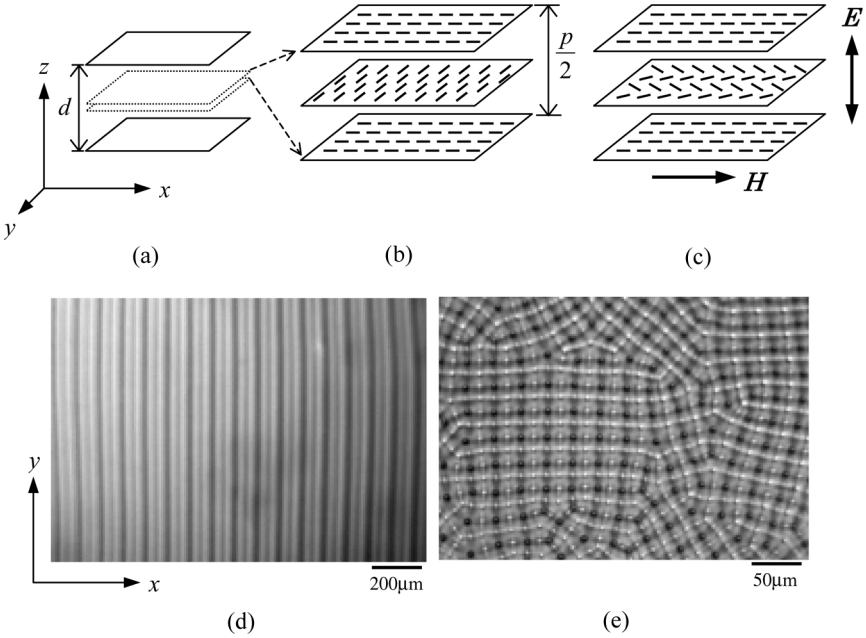


FIGURE 1 Sample cell and typical patterns: (a) the geometry of a sandwich-type cell (CLC), (b) the pitch p of a cholesteric liquid crystal (CLC), (c) the director configuration for applied electric (\mathbf{E}) and magnetic (\mathbf{H}) fields, (d) a Williams domain (WD) observed in nematic liquid crystals (CN_0%), (e) a grid-like pattern (GP) observed in CLCs (CN_3%).

$[\mathbf{H} = (H_x, H_y, 0)]$, as shown in Figure 1(c). At room temperature, investigators observe optical patterns in the xy plane parallel to the glass plates by use of a charge-coupled device (CCD) camera (XC-75, SONY) mounted on a microscope (UNIMAC MS-50, MEIJI TECHNO). For this experiment, in order to capture the patterns with real time on a computer, we used image-processing software (Scion Image Beta 4.0.2) and an imaging board (PCI-VE5, Scion Corporation Company).

First, we investigated the instability and patterns in NLCs as well as CLCs to provide an accurate basis for comparison. In the text and figures the term CN_0% refers to NLCs—pure MBBA without cholesteric dopant CN.

As expected, the NLCs exhibited the well-known Williams domain (WD) pattern, with their corresponding threshold characteristics, as shown in Figure 1(d) and Figure 2, respectively. Certainly, these result from the Carr-Helfrich effect; that is, they were caused by electrohydrodynamic instability due to anisotropy of the liquid crystals

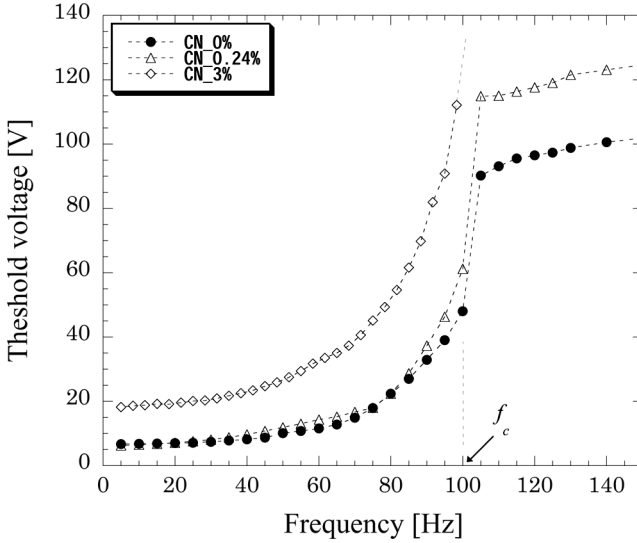


FIGURE 2 Frequency-dependence of threshold voltage for electrohydrodynamic instability. The CN_ c % indicates a mixture of a nematic liquid crystal (MBBA) and a cholesteric dopant (CN) with c %. GPs appear in the CN_3%, while WDs appear in the CN_0% and CN_0.24%. Here, the CN_0% means a pure nematic liquid crystal (MBBA) without the CN.

and periodic charge focusing. Though increasing the applied voltage does produce secondary instabilities [5], this study was limited to investigating the primary instabilities that occur near the threshold and the conductive (low-frequency) regime ($f < f_c \sim 100$ Hz in the present study).

The CLC cells displayed various patterns that varied with the pitch p , as reported in our previous papers [12]. Figure 1(e) shows a pattern typically observed in CLCs. The periodic two-dimensional GPs reflect the characteristics of the CLC's helical pitch. These were clearly different from the WD patterns observed in NLCs, and could be observed in CLCs with a sufficiently small pitch p with respect to d ($p/d \ll 1$). These patterns then appear as a primary instability in CLCs, as shown Figure 2 (see open diamonds; CN_3%), while the WDs appear in the NLC (see solid circles; CN_0%). In case of the CLC cells where $p/d \approx 1$ (CN_0.24%), the grid patterns were not formed.

Next, we investigated the threshold characteristic of the primary instability in the presence of a magnetic field, $H(H_x, H_y, 0)$. We measured the threshold voltages V_{GP} for selected frequencies f in CLCs, and measured V_{WD} in NLCs for comparison [13].

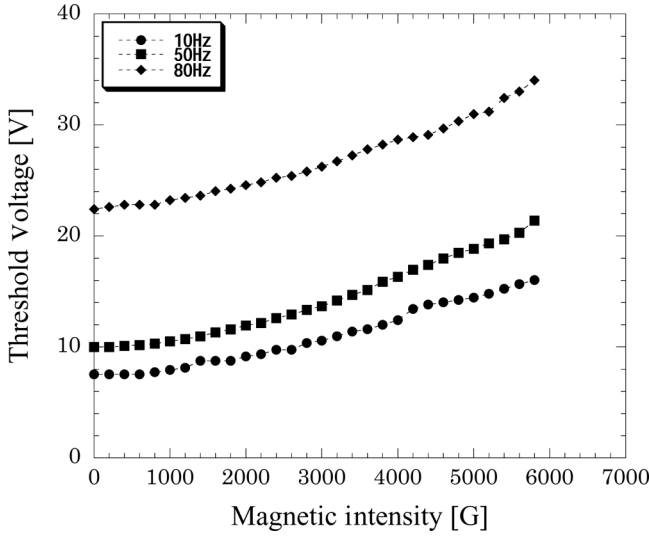


FIGURE 3 H -dependence of threshold voltage for WDs in a nematic liquid crystal (CN_0%).

In case of NLCs (CN_0%), V_{WD} for the WD pattern increases monotonically as H increases, as shown in Figure 3. In case of CLCs, on the other hand, the GPs are found in the presence of H below a certain intensity H^* (~ 2300 G), as shown in Figure 4. Above H^* , however, the WD-like patterns (WDPs; open symbols in Fig. 4), are found at the primary thresholds, instead of the GPs. For $H > H^*$, moreover, the GPs are found as a secondary instability at each selected f , as shown in Figure 4. V_{GP} smoothly increases with increasing $H(>H^*)$. This behavior is very similar to that of NLCs (CN_0%), although the small increase of V_{GP} could not be detected for $H < H^*$.

The unknown WDPs are crucial for the formation of typical GPs in CLCs. Because of the similarity in the optical patterns, it is impossible to distinguish the WDPs (Fig. 5(a)) from the WDs (Fig. 1(d)). However, the WDPs do have some different characteristics. First, the stripes of WDPs always appear parallel to the applied magnetic field \mathbf{H} (i.e., wave vector $\mathbf{k} \perp \mathbf{H}$). In principle, the rolls for WDs are always perpendicular to the reoriented direction of the director \mathbf{n} , which tends toward \mathbf{H} in the case of MBBA with positive anisotropy of magnetic susceptibility χ_a [5,16]. Therefore, the rolls for WDs are perpendicular to \mathbf{H} ($H_x, 0, 0$) for NLC cells with initial director \mathbf{n}_0 (1, 0, 0). Applying a strong magnetic field, \mathbf{H} (0, $H_y \gg H_F$, 0) beyond H_F (~ 1000 G for MBBA) for the twist-Fredericks deformation rotates the WDs around the z -axis. The resulting rolls are then perpendicular to \mathbf{H} ($\mathbf{k} \parallel \mathbf{H}$).

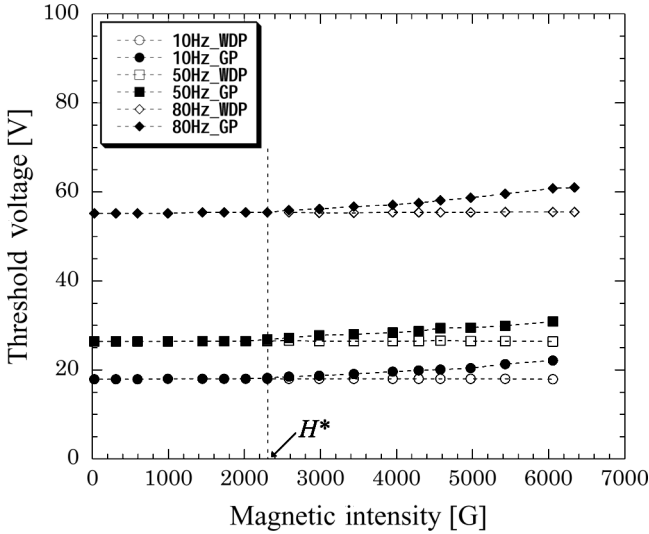


FIGURE 4 H -dependence of threshold voltage for GPs and Williams domain-like patterns (WDPs) in a cholesteric liquid crystal (CN_3%). Solid and open symbols represent GPs and WDPs, respectively, for each selected frequency. See the text for details.

The stripes for WDPs remain parallel to \mathbf{H} ($H_x, H_y, 0$) regardless of the direction in the xy plane ($\mathbf{k} \perp \mathbf{H}$), even for a strong H . For WDPs, however, the typical f -dependence of the threshold is still observed, as shown in Figure 6. Considering the chirality of CLCs [10], these WDP patterns appear to be a result of the electrohydrodynamic (Carr-Helfrich) mechanism.

Furthermore, removing H (or increasing V) causes the WDP to evolve to a GP; however, the WDP disappears if V is eliminated. The magnetic field alone cannot produce the WDPs. This characteristic serves to distinguish the WDPs from similar patterns found in NLCs [14] and CLCs [15] in the presence of H . As shown in Figure 5, when V increases smoothly, the GP (c) evolves from the WDP (a) at a fixed $H_x = 5000$ G. At some point in the process, WDP and grid patterns coexist. Decreasing V smoothly reverses the pattern change ((c) \rightarrow (b) \rightarrow (a)). In addition, the wavelength λ_{WDP} of the WDPs is equivalent to λ_{GP} , such that “Square Grid Patterns” can be found (except for the defects in the GPs seen in Figs. 1(e) and 5(c)). Thus, the GPs must result from a superposition of the stripes of the WDP and of the WD, which are perpendicular to each other.

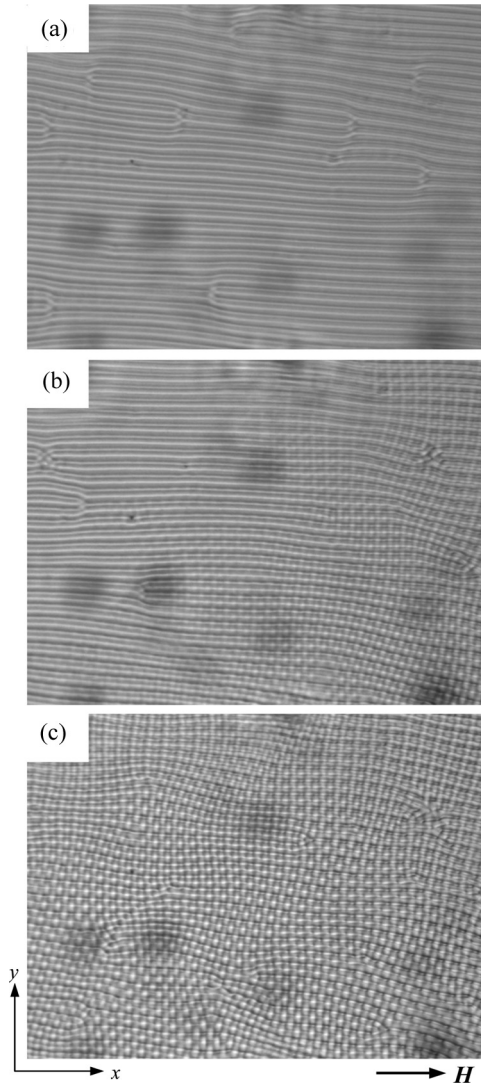


FIGURE 5 Pattern change with increasing voltage V ((a) $V = 27.5$ V, (b) $V = 28.5$ V, and (c) $V = 32.0$ V) for a CLC cell (CN_3%) in the presence of H ($H_x = 5000$ G, 0, 0).

Finally, in the presence \mathbf{H} ($H_x, 0, 0$), we measured the wavelengths for WDs, WDPs, and GPs near the corresponding threshold voltages at a fixed frequency $f = 50$ Hz. In Figure 7, λ_{WD} (for CN_0%) decreases monotonically as H increases, while both λ_{WDP} and λ_{GP} (for CN_3%)

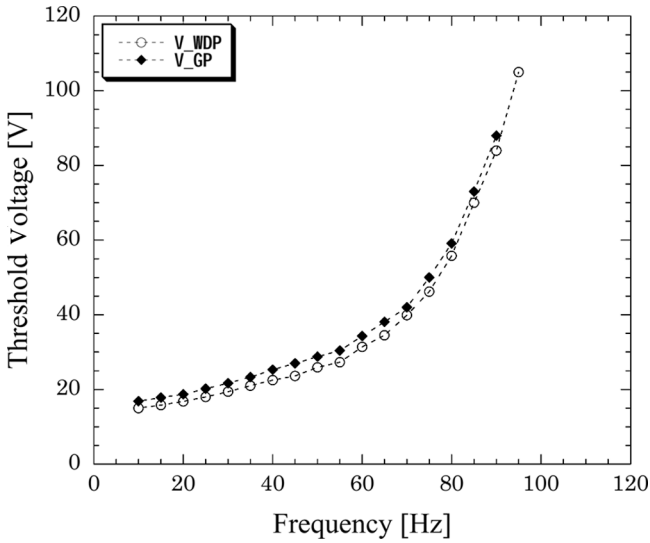


FIGURE 6 Frequency-dependence of the threshold voltages of GPs and WDPs for a CLC cell (CN_3%) in the presence of H ($H_x = 3000$ G, 0, 0).

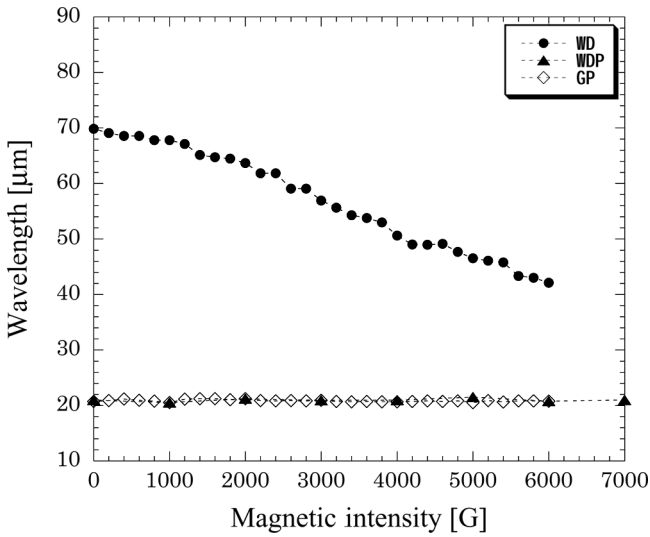


FIGURE 7 H -dependence of the wavelength for WDs, WDPs, and GPs near corresponding threshold voltages. See the text for details.

remain constant with respect to H . In addition, both λ_{WDP} and λ_{GP} are equivalent within our limitation of magnetic intensity ($< H = 7000$ G). In case of NLCs, the wavelength λ_{WD} depends on the effective distance $d_{eff}(H)$, which decreases as H increases. In case of CLCs, however, both λ_{WDP} and λ_{GP} depend on the pitch p [9,12]. We cannot control p in our experiment; therefore, we see no variation of λ_{WDP} and λ_{GP} . The untwisting (critical) intensity H_c is estimated at about 4 T for the present cell (CN_3%) [16]. Accordingly, we expect variations of λ_{WDP} and λ_{GP} beyond $1/2H_c$ [16] and further expect a stepped increase as we increase H [12,16]. We determined that λ_{WDP} varied suddenly beyond $H \approx 3000$ G ($H_c \approx 4400$ G) for CN_0.24%. The p -dependence of λ_{GP} was reported in our previous paper [12].

We do not expect to find WDPs or GPs beyond the untwisting H_c (~ 4 T) because CLCs transform into NLCs at H_c ($p \rightarrow \infty$). The rotation of the stripes of WDPs ($\mathbf{k} \perp \mathbf{H}$) around the z -axis will be also observed near H_c , and then the conventional WDs ($\mathbf{k} \parallel \mathbf{H}$) will be recovered beyond H_c .

SUMMARY

We investigated the characteristics of cholesteric liquid crystals (CLCs) with a controlled helical pitch p . In particular, the threshold behavior for the patterns has been investigated in the presence of a magnetic field H . We found a certain magnetic intensity H^* beyond which Williams domain-like patterns (WDPs) appear as a primary instability. These WDPs appear to be a type of the Williams domains (WDs) induced by electrohydrodynamic instability, with stripes parallel to \mathbf{H} . The WDPs should be distinguished from optically similar patterns without hydrodynamic effects [14,15]. A superposition of the WDP and the WD forms the regular gridlike pattern (GP), which is one of most typical patterns found in CLCs. In the presence of \mathbf{H} , it is possible to understand the wavelength of GPs qualitatively in comparison with that of WDs for nematic liquid crystals.

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