New Characteristics of Electrohydrodynamic Instability in a Nematic Liquid Crystal Doped with a Cholesteric One

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We report electroconvection in a cholesteric liquid crystal with negative dielectric anisotropy, that is, a mixture of p-methoxy benzilidene p-n-butylaniline (MBBA) with a small amount of a cholesteric dopant cholesteryl nonanoate (CN). By changing the corresponding helical pitch by varying the concentration of CN, a variety of ac-driven convective patterns are found in sandwiched sample cells. The pitch and temperature dependences of their threshold voltages and characteristic wavelength of the patterns are systematically investigated.

KEYWORDS: electrohydrodynamic instability, liquid crystal, helical pitch

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In nematics, ac-driven electrohydrodynamic instability has been extensively investigated theoretically and experimentally, and its mechanism and characteristics have been successfully explained.\textsuperscript{1–3} It provides us with a rich variety of stationary and nonstationary patterns. A primary electro-convection, the Williams domain (WD), at a threshold field is found due to the Carr–Helfrich effect.\textsuperscript{4,5} Increasing the electric field and changing its frequency, one also observes various secondary instabilities such as the zigzag instability and the abnormal roll instability.\textsuperscript{6–9} Increasing the electric field further, one observes the well-known dynamic scattering mode (DSM) to be completely turbulent.\textsuperscript{10}

In cholesterics, on the other hand, several groups have also studied ac-driven electrohydrodynamic instability.\textsuperscript{11–18} As in nematics, there is long-range orientational order of the elongated molecules along a local axis described by the director \( n \). In addition, they have an additional orientational order along the helical axis \( h \) in chiral molecules that is always perpendicular to \( n \). In a cholesteric with helical pitch \( p \), by choosing the axis \( h \) along \( z \), the director is given by \( n = (\cos \varphi(z), \sin \varphi(z), 0) \). In principle, one may consider cholesterics (\( p \neq \infty \)) to be a kind of nematics (\( p = \infty \)) with a finite pitch \( p \). However, one may easily expect that the helical order induces noteworthy effects for electrohydrodynamics in the case of a large confinement ratio \( d/p \) (\( > 1 \)) of the standard parallel electrode cells (gap \( d \)). The helical pitch \( p \) must be one of critical factors in understanding electrohydrodynamics and pattern formation in cholesterics.

In the early studies of cholesterics,\textsuperscript{11–18} the threshold field and the spatial characteristics were intensively investigated and successfully explained on the basis of the expanded Helfrich theory for cholesterics.\textsuperscript{19–21} The electrohydrodynamic instability is caused by the competition among the torque induced by Coulomb force, and the elastic and the viscous torque, and then a typical (two-dimensional) periodic pattern appears, similarly to the nematic cases. In theory,\textsuperscript{19–21} the typical two-dimensional pattern periodicity \( \lambda_p \) in cholesterics may be described by \( \lambda_p \sim (p/d)^{1/2} \). With mixing of a cholesteric dopant into nematics, various patterns were investigated a few decades ago.\textsuperscript{15–18}

In this letter we report new experimental results for electroconvection in nematic-cholesteric mixtures for various pitch and/or temperature. In particular, the threshold for the onset of convection and its typical patterns show interesting temperature dependence.

We use a mixture of p-methoxy-benzilidene p-n-butylaniline (MBBA) with a small amount of a cholesteric dopant, cholesteryl nonanoate (CN). We prepare several MBBA–CN mixtures with 0.1 to 5.0 wt% of CN. In this concentration (\( c \)) range, the pitch (\( p \)) varies from 120 to 2.4 \( \mu \)m, which was determined using \( \sigma = 0.12 \pm 0.01 \) m (0 < \( c < 0.05 \)) reported in ref. 14. The experimental investigation is performed on sandwich-type cells whose surfaces are coated with transparent electrodes (indium tin oxide). The electric conductivities and dielectric constants for the present MBBA (at 25\(^\circ\)C) are \( \varepsilon || = 6.50 \times 10^{-8} \Omega^{-1} \text{m}^{-1} \), \( \varepsilon _{\perp} = 3.11 \times 10^{-7} \Omega^{-1} \text{m}^{-1} \), and \( \epsilon || = 4.67, \epsilon _{\perp} = 5.32 \), respectively. The gap between the electrode surfaces with the planar alignment is \( d = 50 \mu \)m and the lateral (active) size is 1 × 1 cm\(^2\).

Across these cells, an alternating electric field \( E = E_0 \cos(2\pi t) \) [\( E = (0, 0, \pm E_2) \)] generated by a function synthesis (NF-1915) and amplified by an amplifier (F20A, Toyo Tech) is applied. The temperature of the cells is stabilized with an accuracy of ±0.2\(^\circ\)C on a hot stage, which is controlled by an electric control system (Digital Controller DB500). The patterns are observed in the xy plane parallel to the electrodes, using a charge-coupled device (SONY XC-75) mounted on a polarizer microscope (ML-9300, Meiji-tech). The applied voltage and the wavelength of patterns are measured with an electric multimeter (Keithley-2000) and an optical micrometer (Nikon), respectively. In order to capture and analyze the patterns on a computer, we have used image process software (Scion Image Beta 4.0.2) and an image board (PCI-VES, Scion Corporation Company).

We have found various patterns in cholesterics at certain voltages \( V \), as shown in Fig. 1. Figure 1(a) indicates the well-known WD at the onset of Carr–Helfrich instability in the 0.1 wt% MBBA–CN cell (C1). The present WD however, has the axis of rolls parallel to the rubbing direction \( n_0 \) for planar alignment (that is, the wave vector \( q \) of the convective mode is perpendicular to \( n_0 \), i.e., \( q \perp n_0 \), \( q \parallel n_0 \)), whereas that in pure nematics is parallel to \( n_0 \)
of this kind of WD (primary WD in Fig. 1(a). Above a certain voltage appears as a secondary instability after the occurrence of the pattern in Fig. 1(b) that shows a chevronlike structure (CV)

We have also found a typical gridlike pattern (GP) in the mixture, which has already been reported in the early studies.11–18) We have observed the GP in a 3.0 wt% MBBA–CN mixture cell, and GP was found in the 3.0 wt% MBBA–CN mixture cell. The scale of the bar in (d) is 50 μm.

This type of evolution (WD or GP) has been reproduced, as predicted in the expanded Carr–Helfrich effect for cholesterics.19–21) How-

In addition, we have found an unexpected interesting pattern in Fig. 1(b) that shows a chevronlike structure (CV) with complicated winding. Upon increasing $V$ it always appears as a secondary instability after the occurrence of the primary WD in Fig. 1(a). Above a certain voltage $V_{\text{CV}}$, winding rolls locally appear in rolls of the WD, and grow widely into a regular CV. The present CV is not stable, but remains throughout the whole (active) area.23–26) Upon increasing $V$ further, we obtain a completely turbulent pattern (DSM), as shown in Fig. 1(d).

We have also found a typical gridlike pattern (GP) in the mixture, which has already been reported in the early studies.11–18) We have observed the GP in a 3.0 wt% MBBA–CN cell (C2, $p = 4.0\,\mu m$) instead of the WD at the onset of convection. It also evolves into the DSM with increasing $V$. This type of evolution (GP → DSM) has been found in other MBBA–CN cells with small pitch [1.0 wt% ($p = 12\,\mu m$), 2.5 wt% ($p = 4.8\,\mu m$), and 5.0 wt% ($p = 2.4\,\mu m$) cells]. The pattern diagrams in the frequency–voltage plane for C1 (with large $p$) and C2 (with small $p$) are shown in Figs. 2 and 3, respectively. In the conduction regime ($f < f_c \sim 450\,\text{Hz}$), the behavior of the well-known primary instability (WD or GP) has been reproduced, as predicted in the expanded Carr–Helfrich effect for cholesterics.19–21) However, WD and CV patterns have been found only in the large-pitch cholesteric cell (C1) for $p > 100\,\mu m$.15–18)

We have systematically investigated the threshold voltage $V_c$ for convection with changing pitch $p$. In order to change $p$, the concentration $c$ of the cholesteric dopant CN was controlled. Figure 4 shows that $V_c$ increases with decreasing $p$ (i.e., with increasing $c$), as expected from elastic energy contributions, for a wide frequency region except high frequencies near $f_c$. From the data in Fig. 4, we see that the relation $V_c \propto p^{-\alpha}$ with $\alpha = 0.58 \pm 0.07$ holds (for $f < 300\,\text{Hz}$), as has been predicted in theory ($\alpha = 0.5$).13,14,19–21) Moreover, $V_c$ diverges at the cutoff frequency $f_c \sim 450\,\text{Hz}$ in all cells. The present $f_c$ is nearly identical in all cells within the range of experimental scatter, since it depends on the conductivity of the host material MBBA.14)

Furthermore, we have investigated the temperature ($T$)-dependence of $V_c$, because the pitch $p$ of cholesterics is sensitive to $T$.27) We observe a strong $T$-dependence of $V_c$ in Fig. 5. Since the other parameters such as elasticity, viscosity, and anisotropies of conductivity and dielectric constant strongly depend on $T$, the direct $p$-dependence of $V_c$ cannot be simply discussed. However, it is worth mentioning that $V_c$ for the GP shows manifestly different $T$-dependence from those for WDs in nematics. As seen in Fig. 5, $V_c$ for GP(A) and GP(B) monotonically decreases with increasing $T$ (even up to $T_c \sim 43\,\text{C}$), whereas $V_c$ for WD(2) in the
Fig. 4. The behavior of the threshold voltage \( V_c \) for GP with changing pitch \( p \) (at \( T = 25^\circ C \)). Here \( p \) was controlled by the concentration of cholesteric dopant CN in MBBA. \( f_c \) represents the cutoff frequency.

Fig. 5. Change of the threshold voltage \( V_c \) with temperature \( T \). Open circles indicate \( V_c \) for GP(A), which is measured with continuously varying \( V \) at various \( T \), and solid circles indicate \( V_c \) for GP(B) with smoothly increasing \( V \) after \( V = 0 \) (in the 3.0 wt% MBBA–CN mixture cell). In order to compare with \( V_c(T) \) for GP, \( V_c \)'s for WDs in pure nematics [WD(1): half-solid squares] and in the 0.1 wt% MBBA–CN mixture [WD(2); crosses] are shown. The applied frequency is fixed at \( f = 100 \) Hz. See the text for further details.

Fig. 6. The wavelength \( \lambda_p \) of GP with respect to pitch \( p \) (at \( T = 25^\circ C \)). \( \lambda_p \) shows the relation \( \lambda_p \propto p^\beta \) with \( \beta = 0.5 \).

Fig. 7. Temperature-dependence of \( \lambda_p \) near onset. \( \lambda_p \) for GP(A) and GP(B) are determined at the corresponding thresholds for GP(A) and GP(B) in Fig. 5, respectively. See the text for further details.

mixture of the 0.1 wt% MBBA–CN diverges at \( T_c \), similar to that for WD(1) in nematics (pure MBBA). This behavior cannot be expected on the basis of the standard theory. In addition, there are two types of temperature properties of \( V_c \) depending on the method of examination. One method is to measure \( V_c \) with varying \( V \) continuously (not removing \( V \)) at various \( T \) [see \( V_c \) for GP(A) indicated by open circles in Fig. 5]. The other is to measure \( V_c \) as follows: for a desired \( T \), set \( V = 0 \) to erase memory effects and increase \( V \) smoothly to determine \( V_c \); repeat the same process for a different \( T \) [see \( V_c \) for GP(B) indicated by solid circles in Fig. 5]. In both methods, the measurement is performed after a sufficient waiting time (about 10 min) to stabilize the state for each desired \( T \). Obviously, the \( T \)-dependence of \( V_c \) (GP) is different for the two cases, while those of \( V_c \) (WD) for nematics and cholesterics are almost the same.

Finally, we have measured the wavelength \( \lambda_p \) of the GP with changing pitch \( p \). \( \lambda_p \) determined near \( V_c \) shows the relation \( \lambda_p \propto p^\beta \) with \( \beta = 0.5 \), as in Fig. 6, which is in quantitative agreement with the theoretical expectation and previous experimental results. Also, the slope \( s = 111.1 \) in Fig. 6 is in good agreement with the theoretical calculation \( s = 3/(2K_{33}/K_{22})^{1/2} = 107.4 \) for elastic constants \( K_{22} = 4.2 \times 10^{-12} \) N and \( K_{33} = 8.61 \times 10^{-12} \) N at \( T = 25^\circ C \) in ref. 30). We have also investigated the \( T \)-dependence of \( \lambda_p \), as shown in Fig. 7. As described above for Fig. 5, \( \lambda_p \) was also determined by the two methods. \( \lambda_p \) for GP(A) shows no dependence of \( T \), while \( \lambda_p \) for GP(B) undergoes a stepped increase with \( T \). Considering that the discontinuous increase \( \lambda_p \) undergoes a stepped increase with \( T \), while \( \lambda_p \) for GP(B) increases with \( T \), and the stepped dependence of \( \lambda_p \) in Fig. 7 seems to be reasonable. Moreover, these aspects in Figs. 5 and 7 may indicate the multistable and subcritical bifurcation properties of the GP states related to GP(A) and (B).

In summary, electrohydrodynamic instability has been systematically investigated in a two-compound mixture of a nematic (MBBA) with a cholesteric dopant (CN). Changing the cholesteric pitch of the mixture, the threshold behavior and the wavelength, as well as the convective patterns, have been studied in detail. In a large-pitch cell, the well-known Williams domain and a chevronlike pattern were found, and their characteristics were described in comparison with the case of pure nematics. The appearance of a one-dimensional
periodic pattern (Williams domain) in a large-pitch cell and a discrete increase of the wavelength for the gridlike pattern (GP) imply that a critical value of the pitch exists for the formation of the two-dimensional periodic pattern (GP).\textsuperscript{15} In other words, in the case of pitch smaller than the critical pitch (with respect to $d$), the Williams domain should be replaced by GP at the onset of convection, due to the superposition and competition of two one-dimensional director modulations. Due to the multistability of GP states, no variation of the GP wavelength was found with temperature change. The twist mode may lead to the subcritical and hysteretic properties leading to multistability. We have qualitatively explained the wavelength characteristics consistently with the theory and previous results.\textsuperscript{11–14,19–21) However, the quantitative explanation for the temperature dependence of the threshold of the GP remains for future investigation.

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23) Such a wavy-like pattern is very similar to the wavy pattern evolved from the preehasy pattern in refs. 24–26, but the origin is completely different.
29) In theory, $V_c$ diverges at $T_c$ for electroconvection in cholesterics as well as in nematics, because the anisotropies $\Delta \varepsilon = \varepsilon_{\parallel} - \varepsilon_\perp$ and $\Delta \sigma = \sigma_{\parallel} - \sigma_\perp$ become zero at $T_c$.