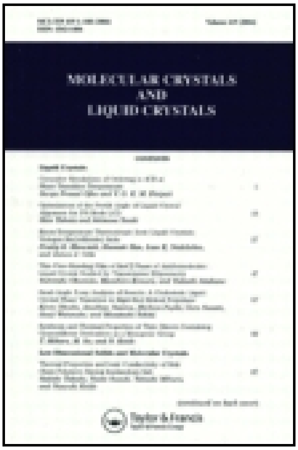

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UNDULATION PATTERNS IN PATTERN FORMATION OF CHOLESTERIC LIQUID CRYSTALS AS WAVELENGTH-CHANGING INSTABILITIES

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The pattern formation in cholesteric liquid crystals (ChLC) is studied in terms of the general pattern selection behaviors near thresholds. Because of the presence of two closely separated thresholds in the ChLC phase, a pattern selection process can be induced in ChLCs without external treatments. It is suggested that, instead of the usual local nucleation, undulation patterns involve the wavelength-changing instability.

Keywords: cholesteric liquid crystal; pattern formation; pattern selection; undulation pattern; wavelength-changing instability

INTRODUCTION

Generally, as nonlinearities of a system grow with system parameters, the current equilibrium state loses its stability and the system moves to a new stable state through fluctuations. This process, known as a bifurcation, has been identified as a useful concept for various phenomena such as phase transitions, chaotic states, and the morphogenesis [1]. This concept from the nonlinear dynamics has been adopted in the study of pattern formation [2]. When a system transfers from a homogeneous state to a patterned one, irrespective of microscopic details, it is well known that the system exhibits a general pattern selection behavior implicit in the amplitude equation [2,3].

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Pattern formations in liquid crystal (LC) phases have attracted much attention since the patterns can be treated as being one-dimensional due to the inherent anisotropy of LC [2]. Among these, the electroconvection of nematic LCs is the most studied case on the basis of the amplitude equation [2–4]. Also, cholesteric liquid crystals (ChLC) have been shown to have various kinds of pattern formation [5–7]. In contrast to the nematic electroconvection, the formed pattern can be maintained in a equilibrium state in ChLC. Thus, the analysis of the patterns in ChLC has been made by starting from the free energy of director distribution in the formed pattern [6]. These ChLC patterns have been considered for use as an optical grating for photonic applications [7]. However, even in the case of the simplest pattern, complicated calculations are involved to get a useful physical insight. Thus, the general interpretation of pattern formation in ChLC has not been made so far.

In this work, we studied the pattern formation in ChLC, without resorting to the elastic free energy of ChLC, but in terms of the general pattern selection concept based on the amplitude equation. We suggest that undulation patterns involve the wavelength-changing instability in the pattern formation of ChLC.

GENERAL PATTERN SELECTION NEAR THRESHOLDS: THE AMPLITUDE EQUATION

In general pattern forming systems, when a parameter of system (control parameter, R), e.g., voltage in ChLC, reaches a certain value (threshold, R_c), pattern formations begin out of the previously homogeneous state with a specific spatial or temporal frequency (critical wavenumber, q_c). Since the nonlinearity which destabilizes the homogeneous state is weak near this threshold, the spatial and temporal modulation of pattern is small in this region. Thus, these modulations can be treated as perturbations of the homogeneous state. Let us assume that a one-dimensional pattern is created perpendicular to $\hat{x}$ in the given coordinate. If the physical quantity varying in this pattern (U), e.g., the direction of director in ChLC, is defined with a complex amplitude (A) as $U(x,t) = [U_0 A(x,t) e^{iq_c x} c.c] + O(\varepsilon)$, the general pattern formation is known to obey the following amplitude equation [2–4]. Here, ε and $c.c$ denote the reduced control parameter: $(R - R_c)/R_c$ and complex conjugate, respectively.

$$\partial_t A = \varepsilon A + [\partial_x - (i/2q_c)\partial_y^2]A - |A|^2 A. \tag{1}$$

The above amplitude equation has a one-dimensional plane wave (coherent pattern) as a time-invariant solution [8].

$$A(x) = (\varepsilon - q^2)^{1/2} e^{iqx} \tag{2}$$

Equation (2) clearly shows that this coherent pattern exists only within the hyperbolic curve (**N**), $\varepsilon = q^2$, while the pattern with the wavenumber outside **N** will disappear into a homogeneous state. This constraint on the existence of coherent pattern highlights some important aspects of general pattern formation phenomena. First, as described earlier, at the threshold of R_c, only the pattern with the critical wavenumber of q_c is observed. Furthermore, the set of allowed wavenumber, the stable q-band, increases with ε. Therefore, it can be deduced that if the control parameter increases above the threshold, the pattern would have various Fourier components, consequently exhibiting a complicated pattern.

However, the amplitude equation has another time-invariant solution which is a unstable saddle-point state, therefore further restricts the stability of coherent pattern (Eckhaus instability) [2,8,9]. The Eckahus instability shows that since the Fourier components outside the curve, $\boldsymbol{E}$(R) = (1/3)· **N**(R) are unstable due to their own stability, these Fourier components should transform to those inside **E** (stable q-band) through a wavelength-changing instability. In the experimental observation, the

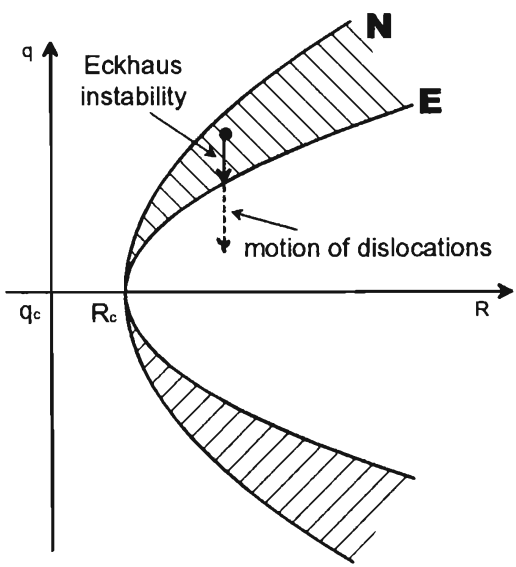

FIGURE 1 The general pattern selection process near a threshold given by the amplitude equation. A unstable Fourier component evolves to the stable q-band via the wavelength-changing instability.

Eckhaus instability usually involves a local nucleation as the wavelength-changing instability [9]. Once the wavenumber crosses the boundary of **E**, the wavelength change is mainly due to the motion of dislocations [2]. Therefore, it can be concluded that a pattern selection process, e.g., the Eckhaus instability, engages wavelength-changing instabilities.

PATTERN FORMATION IN CHOLESTERIC LIQUID CRYSTAL

The pattern formation observed in ChLC phase is discussed based on above concepts. One of the most remarkable things in the pattern formation of ChLC is that the ChLC phase having a positive dielectric anisotropy under a planar anchoring condition has two threshold voltages separated by a few voltages [7].

Let us consider this double threshold structure in detail. In the absence of applied voltage, the ChLC phase has a homogeneous helical structure with the helical axis perpendicular to the planar anchoring surface (**P** state). When the control parameter of ChLC phase, the applied voltage, exceeds a certain thresholds ($V_{\mathbf{P}}$), a pattern formation begins with the critical wavenumber of $q_{\mathbf{P}}$ as expected from the general pattern selection behavior. However, in the ChLC phase, with a large control parameter one can readily obtain another homogeneous state, i.e., in the presence of a high voltage, the field-induced homogeneously homeotropic state is attained (**H** state). Thus, by decreasing the applied voltage below a threshold, another pattern formation can be initiated with the critical wavenumber of $q_{\mathbf{H}}$. In Figure 2, the schematic representation of dual threshold structure is presented. Note the hyperbolic curves at two thresholds which represent the stable region of pattern as given in the preceding chapter by the amplitude equation.

Such dual threshold structure in the ChLC phase was experimentally observed. The ChLC cell with appropriate conditions was fabricated: alignment layers of AL3046 (Japan Synthetic Rubber Co.) were spin-coated on the inner surfaces of cell and both substrates were rubbed unidirectionally to produce a homogeneous planar alignment. Two substrates were assembled with the cell gap of 6.0 μm. The ChLC material was prepared by mixing a LC material, ZLI-2293 (Merck Co.), with a chiral dopant, S-811 (Merck Co., 1.5 wt%, natural pitch: 6.0 μm), and subsequently injected into the cell. The pattern formation in ChLC was observed with an optical microscope with no polarizers as a function of the applied voltage at 1 kHz and the wavelength of pattern was determined from microscopic textures.

Figures 3 show the experimentally observed pattern in ChLC phase. In Figure 3(a) and 3(b) show the growth of pattern and the established

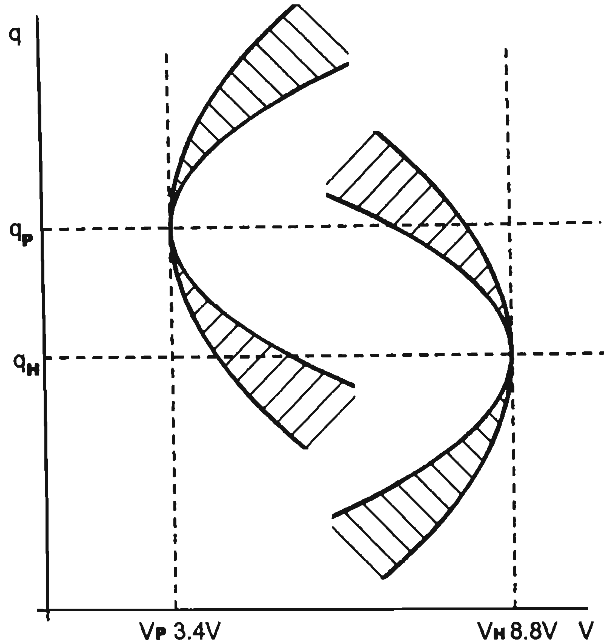

FIGURE 2 The dual threshold structure inherent to the ChLC phase having positive dielectric anisotropy under planar anchoring conditions.

pattern, respectively at the **P** state-pattern transition. The threshold, $V_{\mathbf{P}}$ was 3.4 V and the critical wavenumber, $q_{\mathbf{P}}$ was $2\pi/6.7$ rad/μm. On the other side, the **H** state-pattern transition was initiated at $V_{\mathbf{H}}$ of 8.8 V with the critical wavenumber, $q_{\mathbf{H}}$, was $2\pi/7.2$ rad/μm. Figure 3(c) and 3(d) show the growth of pattern through extensions of cholesteric fingers and the established pattern, respectively. Note that, as already illustrated in Figure 2, two critical wavenumbers, $q_{\mathbf{P}}$ and $q_{\mathbf{H}}$, have quite different values with each other.

This difference in critical wavenumbers can provide one with a way of readily inducing a pattern selection process. Let us assume that a coherent pattern is obtained near the threshold, $V_{\mathbf{H}}$, then this pattern has a Fourier component mainly around $q_{\mathbf{H}}$. Under this circumstance, if the applied voltage is abruptly reduced to just above the threshold, $V_{\mathbf{P}}$, it is highly possible that a certain wavelength-changing instability may occur. This is because the initial critical wavenumber, $q_{\mathbf{H}}$, may not belong to the stable q-band near $V_{\mathbf{P}}$ due to the large difference between critical wavenumbers. Therefore, without any external treatments [9], one may induce the pattern selection process in ChLC phase.

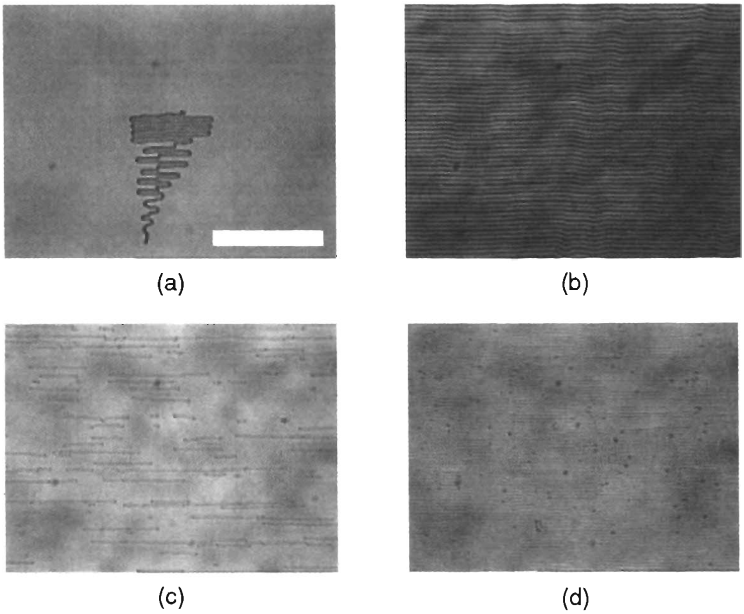

FIGURE 3 The pattern formation in ChLC at V_P: (a) the growth of pattern and (b) the formed pattern. The pattern formation in ChLC at V_H: (c) the growth of pattern and (d) the established pattern. The scale bar in (a) corresponds to 100 μm and all other cases have the same scale.

In the light of above predictions, the pattern selection process provoked by the dual threshold structure of ChLC was experimentally observed. First, a coherent pattern was grown at 8.2 V (slightly below V_H) as shown in Figure 4(a). The main wavenumber was measured as $2\pi/7.4\,\mathrm{rad}/\mu\mathrm{m}$, which was smaller than q_H. Then, in order to induce the pattern selection process, the applied voltage was abruptly decrease to the value near V_P, 3.6 V. Under this circumstance, as shown Figure 4(b), 4(c), and 4(d), the pattern in ChLC exhibited transient undulation patterns which involve the wavelength-changing instability. Once the wavenumbers outside the stable q-band are transformed to those inside the stable q-band, the wavelength-changing instability is mainly due to the motion of dislocations as seen from Figure 4(d) and 4(e). After temporal evolutions, the stable pattern at 3.6 V was developed as shown in Figure 4(e). One more notable thing is that no undulation instability is observed in the reverse change in the applied voltage. When the applied voltage was increased instantly from 3.6 V to 8.2 V, only rapid dislocation motions as shown in Figure 4(f) were observed as the wavelength-changing instability.

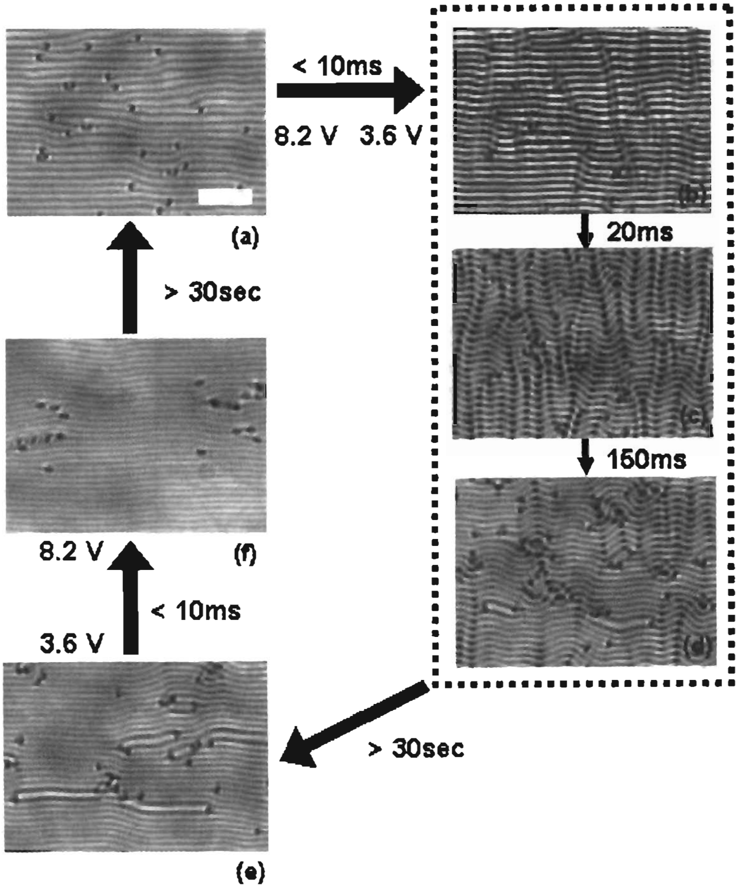

FIGURE 4 The observed pattern formation in ChLC: (a) the coherent pattern grown at 8.2 V, (b)(c)(d) the undulation patterns which engages the wavelength-changing instability, (e) the stable pattern at 3.6 V, (f) the rapid motion of dislocations for changing the wavelength. The scale bar in (a) corresponds to 50 μm and all other cases have the same scale.

Therefore, it may be deduced that, instead of the usual local nucleations, undulation patterns serve as the wavelength-changing instability near the **P** state-pattern transition. Such undulation or zig-zag patterns have been observed in the nematic electroconvection [10]. However, in the nematic electroconvection, the undulated pattern was a stable state, while the undulation instabilities observed in ChLC were a saddle-point state which serves as a wavelength-changing instability. As a summary, in Figure 5,

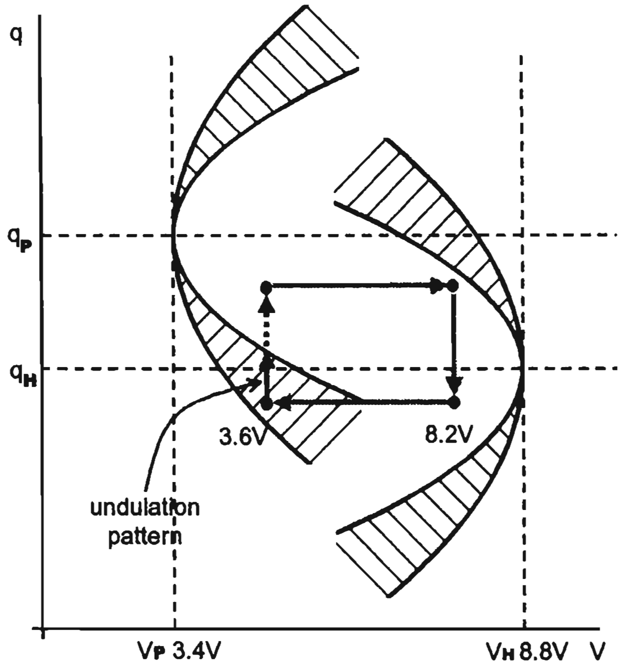

FIGURE 5 The schematic representation of the observed pattern formation in ChLC in terms of the general pattern selection concepts.

we present the qualitative interpretation of the observed pattern formation in ChLC in terms of the pattern selection concepts.

CONCLUSION

We have discussed the pattern formation in ChLC phase in terms of the general pattern selection behaviors observed near the threshold. Due to the two closely separated two threshold structure inherent to the ChLC having a positive dielectric anisotropy with planar anchorings, the pattern selection process can be readily induced without external treatments. In this pattern selection process, it is observed that the undulation patterns act as a wavelength-changing instability near the lower threshold.

As far as we know, the observation of undulation patterns as a wavelength-changing instability has not been reported yet. Naturally, the recognition of undulation pattern as a general wavelength-changing instability like the Eckhaus instability and/or as a saddle-point solution of the amplitude equation has not been made yet. Therefore, if experimental observations and theoretical studies on undulation patterns are carried out further, many interesting aspects of pattern formation may be revealed.

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