Prewavy instability of nematic liquid crystals in a high-frequency electric field
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A kind of electrohydrodynamic instability, the prewavy instability, in nematic liquid crystals is reported. The characteristic of the instability was optically investigated and discussed in comparison with some similar instabilities. Obviously the instability partially shows an isotropic feature around the nematic-isotropic transition temperature. Owing to the characteristic properties of the flow and the spatial periodicity, it should be distinguished from a previously proposed isotropic instability.

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I. INTRODUCTION

ac-driven electrohydrodynamic instability in liquid crystals is one of the prime examples of the physics of pattern formation in dissipative systems out of equilibrium. It provides us with a rich variety of stationary and nonstationary patterns that have been intensively studied for the last three decades [1,2]. A primary electroconvection, Williams domain (WD), at a threshold field is found due to the so-called Carr-Helfrich instability [3–5]. Increasing the electric field and changing its frequency, one also observes various secondary instabilities such as the zigzag instability, the Eckhaus instability, the abnormal roll instability, and so on [6–10]. In the present paper we report a kind of instability called the prewavy instability [11] observed in a nematic liquid crystal, and discuss its characteristic in comparison with some similar instabilities [12–18]. Though the prewavy and similar instabilities were already observed and discussed in experiment and theory [13,15–19], open questions and discrepancies still remain. Well-ordered stripe patterns such as the present prewavy pattern were optically observed slightly above the threshold field. In fact the optical stripe patterns due to the prewavy instability may be very similar to wide domains formed by the anisotropic inertia mode instability reported before [14,15]. In contrast to this, observation of mass flows in the prewavy pattern appear the nematic-isotropic transition temperature shows a similarity with an isotropic electrolyte mode [12,16]. Sometimes, this isotropic mode cannot be distinguished from the anisotropic dielectric mode observed above a certain high frequency \( f_c \), i.e., cutoff frequency [12,16]. From these reasons the prewavy instability, which is not well understood yet, might be confused with abovementioned instabilities.

The aim of this paper is to clarify the difference of the prewavy instability from other similar instabilities, and to propose a possible mechanism based on experimental results. We argue about the main features of the prewavy instability, i.e., the frequency dependence on the threshold voltage with varying the electric conductivity, the voltage and the frequency dependence on the characteristic wavelength, the geometry of the flows, and the behavior of the threshold voltage around the nematic-isotropic transition temperature.

II. EXPERIMENT

The experimental investigation was performed on sandwich-type cells whose surfaces were coated with transparent electrodes (indium tin oxide). The electric conductivity of a nematic liquid crystal MBBA (\( p \)-methoxybenzilidene-\( p' \)-n-butylaniline), was controlled by doping with TBAB (tetra-\( n \)-butyle-ammonium bromide). An alternating electric field \( E = E_0 \cos(2\pi ft) \) was applied perpendicularly to the electrodes \( E = (0,0, \pm E_0) \). The thickness \( d = 20–250 \) \( \mu m \) of a nematic slab with lateral (active) size \( 1 \times 1 \) \( cm^2 \) was controlled by a polymer spacer (Mylar). The temperature of the cells was stabilized with an accuracy of \( \pm 0.2 ^\circ C \) in a hot stage, which was controlled by an electric control system (Digital Controller DB500). The patterns were observed in the \( xy \) plane parallel to electrodes by use of a charge-coupled-device camera (SONY XC-75) mounted on a polarizer microscope (Nikon). In order to investigate flows, small particles [Micropearl (Sekisui Chemical) of diameter 3.88 \( \mu m \)] were introduced into liquid crystal cells. The threshold voltages and the wavelength of patterns were measured with an accuracy of \( \pm 5 \% \) with an electric multimeter and an optical micrometer, respectively.

III. RESULTS

A. The optical characteristics

A typical prewavy pattern appears in a homeotropic alignment cell, as shown in Fig. 1(a). It is well ordered with a characteristic spatial period \( \lambda_w (>4d) \) much larger than the normal convective roll period \( \lambda_{WD} \). It is also characterized by the director modulation in the \( xy \) plane (due to the in-plane rotation of the director), so that one can distinguish it from a WD with the director modulation in the \( xz \) plane. The in-plane rotation of the director was developed by the prewavy instability following the Fréedericksz transition (at 3.5 V). For the sake of convenience we define the \( x \) axis parallel to an in-plane orientation of the director in the Fréedericksz state, which is spontaneously selected in the homeotropic cell. In the prewavy patterns the in-plane rotation angle \( \alpha \) of the director with respect to the \( x \) axis \( (\alpha = 0) \) increased with increasing voltage \( V (> V_w) \); see below).
saturated to a maximum angle \( \alpha_{\text{max}} (\sim 45^\circ) \). The detail of \( \alpha(V) \) was reported in our previous paper [21]. Only due to the in-plane rotation of the director, no prewavy pattern is detectable without two polarizers. The cross polarizers give us the best contrast of the prewavy pattern, as seen in Fig. 1(a), while it remains visible with parallel polarizers [22].

Increasing voltage at a fixed frequency \( f > f_w \) (see below), one can observe a chevron pattern, defect-free chevron [DFC, Fig. 1(b)] [23], characterized by a double periodicity. The DFC is also a typical pattern, in which electroconvective rolls (WD) are superimposed on the prewavy pattern [11,22]. Notice also that the alternating zig and zag rolls of the DFC always form their axes normal to the corresponding in-plane directors, as seen in Fig. 1(b). This chevron pattern is detectable in one or two polarizer(s) and even removing them.

**B. The frequency dependence of the threshold voltage**

The frequency dependence on the threshold voltage of the prewavy instability was investigated in three (homeotropic) cells with different electric conductivity \( \sigma \), because the threshold of such an electric instability usually depends on \( \sigma \). Figure 2 shows the threshold lines (solid circles) of the prewavy pattern above a certain characteristic frequency \( f_w \). The threshold \( V_w \) of the prewavy instability weakly depends on \( f \) in all cells [20]. Near \( f = O(\tau^{-1}) \sim 180 \text{ Hz} \) in cell 1 with low \( \sigma \) when increasing \( V \), there appears a dielectric chevron (DC) instead of the DFC at a threshold voltage after appearance of the prewavy patterns. Here \( \tau_{\sigma} \) corresponds to the charge relaxation time (\( \sim 10^{-2} \text{ s} \)). In Fig. 2, moreover, the prewavy pattern begins to appear in lower \( V \) and higher \( f \) with the increase of \( \sigma \).

**C. The periodicity with respect to frequency and voltage**

When a characteristic wavelength \( \lambda_w \) of the prewavy pattern was defined in Fig. 1(a), \( f \) and \( V \) dependence of \( \lambda_w \) was measured, as shown in Fig. 3. \( \lambda_w(f) \) at a fixed \( V = 60.1 \text{ V} \) and \( \lambda_w(V) \) at a fixed \( f = 2100 \text{ Hz} \) are almost constant within experimental scatters. Here \( \lambda_w \) is around 6\( d \). No change of \( \lambda_w(f) \) was confirmed in a range from \( f = 1500 \text{ Hz} \) to 10 kHz. \( \lambda_w(V) \) was also independent of \( V \).
below a certain threshold voltage $V_{\text{wavy}}$ at which it evolved to a sinusoidal wavy pattern with remarkable change of $\lambda_w$ [20]. For all cells of planar and homeotropic alignments with different $d$, no $f$ and $V$ dependence of $\lambda_w$ were observed below $V_{\text{wavy}}$. $\lambda_w$ was always measured in the range of $5 \pm 1d$.

D. The flows accompanied with the instability

Since the flow is important to understand the mechanism of electrohydrodynamic instabilities, it was carefully investigated by introducing small particles into a cell. The flow optically observed in the prewavy pattern showed three figures with respect to the trajectory of the particles. (1) The particles moved along the director orientation, that is, they moved zigzag cross the stripes, as shown in Fig. 4. And the flow with the opposite direction was also observed on a different focus plane. Obviously, this is a main flow in the prewavy pattern. (2) Sometimes some of them moved in mutually opposite directions along the stripes on a same focus plane. It seemed to be a long circular flow in the $xy$ plane. (3) Rolling and rotating of a lump of particles was often observed. The particles also moved at voltages below the threshold $V_w$ of the prewavy instability. In this case their trajectory was not totally random and lay along the $x$ direction with large scatters as shown in Fig. 5(a). Above $V_w$ the velocity of the $y$ direction $v_y$ is clearly split as seen in Fig. 5(b). This means that the particles move zigzag cross the stripes in the prewavy pattern, that is, they move along the director modulated periodically by the prewavy instability.

The feature of the main (zig and zag) flow in the prewavy pattern was quantitatively investigated. We defined an angle $\beta$ characterizing the flow direction, i.e., an in-plane angle between the flow direction and the $x$ axis. The investigation was carried out with observing the motion of 50 particles that were selected randomly in a cell. The particles started to move about 55 $V$ ($V_f$ in Fig. 6). The averaged angle $\bar{\beta}$ was measured as a function of $V$ in Fig. 6. The $\bar{\beta}(V)$ curve indicates a bifurcation. Above $V_w$ ($\sim 84 V$) the flow profile seems to be regular and shows zig and zag flow. Below $V_w$, one can see that $\bar{\beta}=0$ and has larger standard deviation. The behavior of $\bar{\beta}(V)$ and its amplitude on $V$ are very similar to $\alpha(V)$, the in-plane rotation angle of the director (see Ref. [21]). This means that above $V_w$ the direction of the flow is always parallel to the in-plane director in the prewavy pattern.

E. The temperature dependence of the threshold voltage

The temperature-dependence of $V_w$ was investigated with increasing temperature $T$. By measuring the threshold value of the optical pattern and the prewavy flows simultaneously, $V_w$ was determined with an accuracy of $\pm 5\%$. Figure 7 shows that $V_w(T)$ decreases monotonously to the nematic-isotropic temperature $T_c$ ($\epsilon_f=0$). Here $\epsilon_f=\epsilon_f(=T-T_c)$ is a relative temperature from $T_c$. Then the optical (prewavy) pattern disappeared just beyond $T_c$, but the flow still remained. Obviously, $V_w(\epsilon_f<0)$ continues to the threshold of the flow in the isotropic phase ($\epsilon_f>0$) without abrupt change. In this isotropic phase ($\epsilon_f>0$) a lump of the particles observed in the prewavy pattern ($\epsilon_f<0$) still rolled...
and rotated, and almost of the single particles moved randomly in the xy plane. On the other hand, the thresholds of WD, DFC, and DC diverge at $T_c$, because they are all anisotropy-originated modes in the nematic phase.

In addition, the frequency dependence of $V_w$ in the isotropic phase ($\varepsilon_f < 0$) was measured, in order to compare with $V_w(f)$ in the nematic phase ($\varepsilon_f > 0$). It shows a plateau at low frequencies and smooth increase of $V_w$ with increasing $f$, as seen in Fig. 8. This behavior of $V_w(f)$ does not change below and above $T_c$. In this measurement $V_w(f)$ below $T_c$ was determined by the optical pattern and the flow, while it was done only by the flow (rolling or rotating) above $T_c$.

**IV. DISCUSSION AND CONCLUSION**

In order to classify the prewavy instability observed at a high frequency regime ($f > f_w$), it should be compared to the previously proposed ones such as the dielectric, inertia, and isotropic (electrolyte) modes that were also observed in high frequencies. There are a lot of similarities between the prewavy and inertia mode with regard to the optical patterns and the frequency dependence of the threshold voltage, as described Secs. III A and III B. However, the inertia as well as the dielectric mode was known as an anisotropic mode that could appear only in the (anisotropic) nematic phase [15,16]. Their modes always show a distinct divergence at the nematic-isotropic transition temperature $T_c$, as shown in Fig. 7 (see DC) and Fig. 7 in Ref. [15]) while the prewavy instability continues around $T_c$. In addition, no dependence of the prewavy wavelength ($\lambda_w$) with respect to voltage and frequency in Fig. 3 also distinguishes the prewavy instability from the dielectric, inertia, and isotropic (electrolyte) modes [25]. We summarize these differences in Table I to compare easier.

On the other hand, the prewavy instability shows some difference from the isotropic (electrolyte) mode proposed previously [12,16,25], though both have a continuity of the threshold of the flow around $T_c$. Compared to Refs. [12,13,16], the most different feature is the trajectory of the flows. According to their isotropic mode, the vortices of the particles were a main flow in nematic phase with their
size of order of cell thickness, but no zigzag and long circular flows [(1) and (2) of the prewavy flows in Fig. 4] were observed. Moreover, no (isotropic-type) wide stripe pattern such as the present prewavy pattern has been reported. Thus, the prewavy instability should be classified as a different type of isotropic mode. In case of the previous isotropic mode, the prewavy instability should be classified as a different type of isotropic mode. In case of the previous isotropic mode, however, it is impossible to expect our large prewavy domains and the characteristic flow in the prewavy pattern.

In conclusion, the present prewavy instability should be classified as a different type of isotropic mode distinguishable from the other high-frequency modes. Here a coupling model between the isotropy-originated flow described above and the director might be considered to understand the prewavy instability. Namely, the strongly developed flow at high voltages could induce an inhomogeneous (twist) Frenedericksz transition (e.g., see Ref. [26]), i.e., the prewavy instability. Moreover, the large wavelength of the prewavy patterns could be formed by the elastic constants and viscous coefficients of the liquid crystal [26]. The wavy and spiral wavy patterns successively evolved from the prewavy pattern are also an interesting subject to understand the nature of pattern formation in dissipative systems [19,20].

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### Table I. Comparison between the present prewavy mode and other similar modes at high frequencies.

<table>
<thead>
<tr>
<th>Mode</th>
<th>Dielectric mode</th>
<th>Inertia mode</th>
<th>Isotropic mode</th>
<th>Prewavy mode</th>
</tr>
</thead>
<tbody>
<tr>
<td>$V_{th}(f)$</td>
<td>$f^{1/2}$</td>
<td>$f^{1/2}$</td>
<td>$f^{1/2}$</td>
<td>$f$</td>
</tr>
<tr>
<td>$\lambda_{th}(f)$</td>
<td>$f^{-1/2}$</td>
<td>$f^{-1/2}$</td>
<td>$f^{-1/2}$</td>
<td>$f^{-1/2}$</td>
</tr>
<tr>
<td>$V_{th}(V)$</td>
<td>$-3d(V&lt;V')$, $-3d(V&gt;V')$</td>
<td>$V_{th}(T=T_c)$</td>
<td>$V_{th}(T=T_c)$ = $V_c(f)$</td>
<td>$V_{th}(T=T_c)$ = $V_c(f)$</td>
</tr>
<tr>
<td>Flow</td>
<td>small surface flow</td>
<td>long circular flow</td>
<td>convective flow</td>
<td>zig-zag flow, long circular flow, rolling/rotating flow</td>
</tr>
</tbody>
</table>

*Also $\lambda_{th}(V) = \lambda_{th}(V=V_{th}) - a V$ in Ref. [11].*
[23] In order to distinguish the chevron pattern in Fig. 1(b) from a defect-mediated chevron, we named the defect-free chevron (DFC) for Fig. 1(b). See Ref. [11] for the details.