Observation and determination of abnormal rolls and abnormal zigzag rolls in electroconvection in homeotropic liquid crystals

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(Received 9 January 1998; revised manuscript received 6 July 1998)

Direct evidence for two different types of normal rolls and of zigzag rolls in homeotropically aligned nematic liquid crystals in a magnetic field is reported. The conventional normal rolls have the reflection symmetry in the xy plane. The instability, however, breaks the reflection symmetry $y \rightarrow -y$ on the director and then the abnormal rolls are expected to be observed. We have investigated the instability experimentally and discussed it in terms of the recent numerical results by Plaut *et al.* [Phys. Rev. Lett. **79**, 2367 (1997)]. Due to the new instability, the abnormal zigzag rolls are also found below the Lifshitz frequency.
[S1063-651X(98)05012-0]

PACS number(s): 47.20.Lz, 47.20.Ky, 47.65.+a

I. INTRODUCTION

The electrohydrodynamic instability in planarly aligned nematic liquid crystals has been actively studied as an attractive pattern formation system, where the primary pattern appears as periodic rolls. In the conductive regime one can observe normal rolls (NRs) with the wave vector **k** of the convection rolls parallel to the preferred direction $\mathbf{n_0}$, defined as the x axis, beyond the Lifshitz frequency f_L, and oblique rolls (ORs), which show a certain angle between **k** and $\mathbf{n_0}$ below f_L, respectively [1–8].

Very recently, an instability in the planar system, which corresponds to a homogeneous in-plane distortion of the director **n** in the xy plane, was reported [9]. This instability breaks the reflection symmetry $y \rightarrow -y$ for the director and then abnormal rolls (ARs) are observed. For ARs, **k** is parallel to $\mathbf{n_0}$ but the **C** director is not parallel to $\mathbf{n_0}$, so there is a small angle between **k** and the **C** director where the **C** director represents the projection of the director **n** onto the xy plane. Plaut *et al.* have predicted the existence of ARs by a nonlinear analysis including an additional mode, i.e., the twist mode corresponding to a homogeneous rotation of the director in the xy plane [9,10].

It is well known that configurations that imply an azimuthal rotation of the director can be determined using a cross-polarizer set [1,2]. However, since in the planar system the director **n** is fixed to the x axis at the two electrodes, the azimuthal rotation of directors is accompanied by a twist deformation along the z axis. Therefore, the polarization of the incident light follows adiabatically the twisted director and leaves the cell without any rotation. Consequently, since the transmitted light has the same polarization as the incident light, the twisting orientation of the directors in the planar system is not detectable with a cross-polarizer set, although the domain boundaries can be simply observed (the present authors do not discuss the domain boundaries, but the domains themselves with different twist orientation). Fortunately, it is known that the additional insertion of a quarter wave plate to a cross-polarizer set raises the degeneracy between clockwise and counterclockwise twist domains and allows the twist deformation of the director to be detectable in the planar system [11,12]. Then one can observe a clear optical contrast between the two domains with the director twisted in opposite directions [13].

In the present paper we report the existence of two types of normal roll patterns and two types of zigzag patterns, respectively, which occur as instabilities in a homeotropically aligned system in a magnetic field parallel to the electrodes ($\mathbf{H}=H\hat{x}$). In the homeotropic system the director tilted by the bend Fréedericksz transition is allowed to free azimuthal rotation in the plane without a magnetic field. When a magnetic field is applied to the system, the rotation is suppressed, similarly to the effect of the anchoring force by boundaries in the case of the planar system [7,14–19]. Accordingly, when the in-plane distortion is induced, the present homeotropic system may be regarded to be similar to the planar system, except that it has no twist deformation along the z axis. Therefore, in the case of the homeotropic system in a magnetic field, the in-plane distortion of the director must be detectable in the cross-polarizer set without a quarter wave plate [13] because the twist deformation does not exist in the system. However, the conventional optical observation for the most frequently observed electroconvection patterns has always used a polarizer for the extraordinary light that monitors only tilt profiles of the director in uniaxial nematic liquid crystals [20]. Consequently, although Richter *et al.* had proposed *abnormal rolls* [21], to our knowledge, no detailed study has been done so far. We have investigated the instability related to the appearance of ARs not only in the NR regime but also in the OR regime for the homeotropic system in a magnetic field.

II. EXPERIMENT AND DISCUSSION

We use the nematic liquid crystal MBBA (p-methoxybenzilidene-p'-n-butylaniline), which is filled between two parallel glass plates whose surfaces are coated with transparent electrodes, indium tin oxide. The distance $d=52$ μm between the glass plates is maintained with polymer spacers and the lateral size of the system is 1×1 cm^2. The details of the sample cells and experimental setup were already described in Ref. [15]. The constant magnetic field is applied parallel to the glass

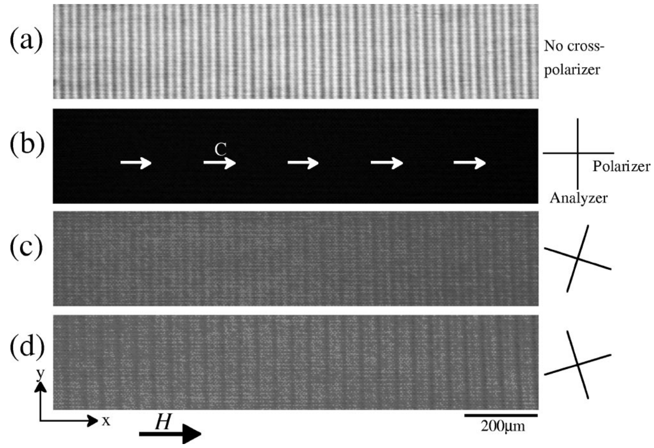

FIG. 1. Experimental observation of the normal rolls ($H_x = 1600$ G, $f = 2000$ Hz, $V = 14.48$ V) (a) without a cross-polarizer set, (b) with a cross-polarizer set whose polarization of the polarizer is parallel to **H**, (c) with a 15° clockwise rotation of the cross-polarizer set, and (d) with a 15° counterclockwise rotation of the cross-polarizer set. All pictures are obtained from sufficiently large two-dimensional images. The white arrow in (b) represents the **C** director in the *xy* plane. The **C** director well uniformly aligns parallel to **H** in the normal rolls.

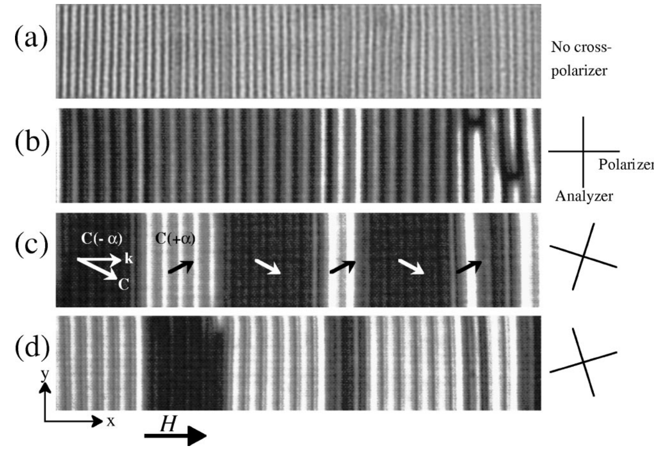

FIG. 2. Experimental observation of the abnormal rolls ($H_x = 1600$ G, $f = 2000$ Hz, $V = 14.86$ V) (a) without a cross-polarizer set, (b) with a cross-polarizer set whose polarization of the polarizer is parallel to **H**, (c) with a 15° clockwise rotation of the cross-polarizer set, and (d) with a 15° counterclockwise rotation of the cross-polarizer set. The arrow in (c) represents the **C** director in each domain of the *xy* plane. The domains with the different **C** director in the abnormal rolls can be clearly distinguished.

plates ($H_x = 1600$ G) and the frequency f of the applied electric field is fixed at 2000 Hz for NRs and 100 Hz for ORs, respectively ($f_L \simeq 600$ Hz at $H_x = 1600$ G).

For the NR regime ($f = 2000$ Hz) Fig. 1(a) shows the conventional NR at a voltage $V_N = 14.48$ V without a polarizer, which is characterized by a wave vector **k** of the convection rolls parallel to the preferred direction **H** (playing a role similar to $\mathbf{n_0}$ in the planar system). First, inserting into the optical apparatus the cross-polarizer set such that the incident light becomes linearly polarized parallel to **H** (i.e., the x axis), we could clearly determine the orientation of the **C** director because the minimum optical intensity is expected in the case of the **C** director parallel to the polarization plane of a polarizer [21]. As shown in Fig. 1(b), **C** directors of NRs are parallel to **H** (or **k**) in the whole two-dimensional plane. When we define an angle α between the **C** director and **k**, NRs are characterized by $\alpha = 0$. In addition, rotating the cross-polarizer set around the z axis, we observed that only the optical intensity of the whole two-dimensional pattern has homogeneously changed, as shown in Figs. 1(c) and 1(d), since the orientation of the **C** director was uniformly ordered by **H**.

On the other hand, we observed the other type of normal rolls, i.e., the ARs at a voltage V_A ($= 14.86$ V) larger than V_N, as shown in Fig. 2. However, one cannot distinguish ARs in Fig. 2(a) from NRs in Fig. 1(a) without the cross-polarizer optical set. By rotating the cross-polarizer set, the orientation of the **C** director can be determined as shown in Figs. 2(c) and 2(d). The maximum contrast has been obtained when rotating the cross polarizer by an angle of 15° either clockwise or counterclockwise. The **C** directors of ARs are indicated by black and white arrows in Fig. 2(c), which are determined from optical intensity changes. The angle α is clearly finite in ARs ($\alpha \simeq \pm 15°$) in contrast to the case in NRs ($\alpha = 0$). There coexist clearly visible, well separated domains with $\mathbf{C}(+\alpha)$ directors and $\mathbf{C}(-\alpha)$ directors in ARs, as can be seen in Fig. 2(c).

Moreover, we observed two types of transitions, i.e., a direct transition from NRs to ARs, and one from NRs to ARs via a zigzag instability, for frequencies above and below a certain characteristic frequency [$f_{AR} \simeq 1100$ Hz at $H_x = 1600$ G (ω_{AR} of Fig. 3 in Ref. [9])], respectively. The experimental results obviously show that ARs are accompanied by the $y \to -y$ symmetry breaking on the director in the homeotropic system in a magnetic field. Because ARs in the present study can be observed by a rotation of the cross-polarizer set without a quarter wave plate, it is obviously different from the one related to the twist deformation in the planar system.

The investigation of the orientation of the **C** director in OR regime ($f = 100$ Hz) was carried out with the same experimental procedure. First, two different types of zigzag patterns, which are called *Z*-I and *Z*-II hereafter, were observed. Figure 3(a) shows a zigzag pattern (*Z*-I) at $V_{Z-I} = 8.40$ V. Inserting and rotating the cross-polarizer set, it was proved that in *Z*-I the **C** directors were uniformly parallel to **H** in the whole area, i.e., the direction was similar to NRs described above. When the applied voltage was increased to $V_{Z-II} = 8.99$ V, a different zigzag pattern (*Z*-II) was observed, as shown in Fig. 3(b). *Z*-II obviously consists of two types of domains with different angles of **C** directors with respect to **H**, of which a zig and a zag have similar orientation to ARs in Fig. 2(c). For *Z*-II, therefore, rotating the cross-polarizer set by an angle of 15° either clockwise or counterclockwise, a maximum contrast is obtained. Thus the angle between the **C** director and **H** can be estimated as about 15°, which is roughly the same angle as that of ARs. Increasing the voltage further, zig and zag rolls become more like normal rolls ($\pm k_y \to 0$), where the domains with different **C** directors in *Z*-II persist. Although **k** becomes parallel to **H**, the **C** director is not parallel to **H**. Therefore, α remains nonzero in analogy to ARs.

To validate the change from *Z*-I to *Z*-II in the OR regime,

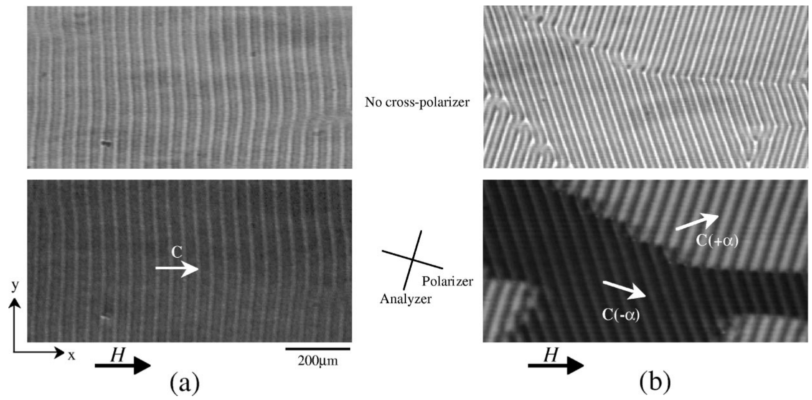

FIG. 3. Two types of oblique rolls ($H_x = 1600$ G, $f = 100$ Hz). (a) Zigzag patterns (Z-I) with uniform orientations of the **C** director in the whole xy plane are obtained at $V = 8.40$ V. (b) Zigzag pattern (Z-II) with distinct domains of the different **C** director in the xy plane at $V = 8.99$ V. The upper pictures are observed without a cross-polarizer set and the lower ones under a 15° clockwise rotation of the cross-polarizer set, respectively. The white arrow in the lower pictures represents the **C** director in the xy plane.

we measured the dependence of α on the control parameter defined as $\varepsilon = (V^2 - V_c^2)/V_c^2$, where V_c is a critical voltage. Figure 4 shows that the azimuthal rotation α of the director develops only for $\varepsilon \geqslant 0.05$ and that it saturates to $\alpha_{sat} \simeq 35°$ at $\varepsilon \simeq 0.25$. Thus we were led to define a transition point $\varepsilon_{ZI-II} = 0.05$. The obliqueness β for the rolls is also measured with increasing ε. β starts to increase at ε_{ZI-II} ($\simeq 0.05$) as α does, that is, α and β simultaneously start to increase with the increase of ε. However, first β shows a saturation towards $\beta_{max} \simeq 12°$ at $\varepsilon_{\beta_{max}} \simeq 0.1$ before it shows a decrease with an increase of ε, while α continues increasing with increasing ε. Then ARs with α_{sat} and $\beta = 0$ appear at $\varepsilon_{AR} \simeq 0.5$. This behavior may indicate the independent behavior of α and β on ε from each other. That is, below ε_{ZI-II} (i.e., the Z-I region) the zigzag rolls appear with $\alpha = 0$, while above ε_{ZI-II} (i.e., the Z-II region) the zigzag rolls have finite α and β. Therefore, we concluded that the zigzag rolls must be Z-I at $0 \leqslant \varepsilon \leqslant \varepsilon_{ZI-II}$ and Z-II at $\varepsilon_{ZI-II} < \varepsilon \leqslant \varepsilon_{AR}$. The transition point from Z-I to Z-II depends on the magnitude of the applied magnetic fields H as well as the frequency f of the applied voltage.

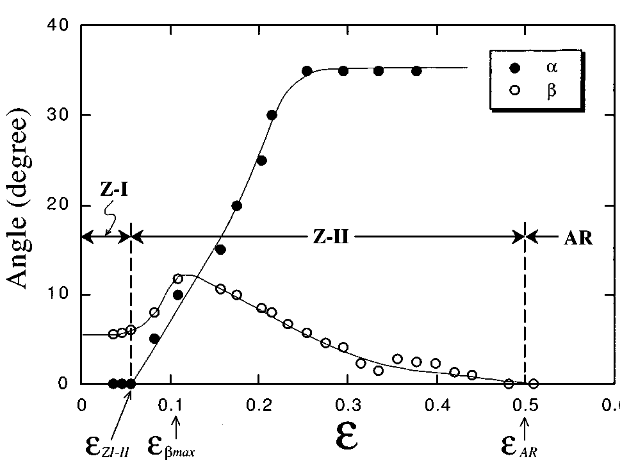

FIG. 4. α and β dependences on applied voltages in the OR regime ($H_x = 1600$ G, $f = 100$ Hz). α indicates the rotation angle of the **C** director against H, β the obliqueness for the rolls. The thin lines are a guide for the eye. There are two characteristic voltages (i.e., ε), one ε_{ZI-II} ($\simeq 0.05$) for the change from Z-I to Z-II and the other ε_{AR} ($\simeq 0.5$) for the change from Z-II to ARs (see the text).

In both the NR and OR regimes the threshold ε_α for the azimuthal rotation of the director increases with increasing H, which tends to suppress the instability. The summary of the pattern changes is given in Fig. 5. The horizontal lines drawn schematically indicate the regions where the patterns are observed. The AR in Figs. 5(b) and 5(c) corresponds to R-normal rolls in Fig. 12 of Ref. [14]. The experimental results obtained in the NR regime of the homeotropic system are in qualitatively good agreement with the numerical results in Ref. [9]. However, the calculations in this article were done based on the planar geometry and unfortunately the homeotropic alignment was not considered. With the instability being induced, there should be an intrinsic difference between the two systems ($\partial\alpha/\partial z = 0$ for the present homeotropic system, but $\partial\alpha/\partial z \neq 0$ for the planar one). Therefore, no prediction of the change from Z-I to Z-II in the OR regime exists in Ref. [9].

The destabilizing mode for the in-plane distortion (i.e., a uniform azimuthal rotation of the director) in the present homeotropic system may be induced by nonlinear coupling between the director field and the velocity field and then the azimuthal rotation angle α is reinforced by these two fields [9,12]. This may induce the instability due to the in-plane distortion.

It is worth mentioning the relationship between the

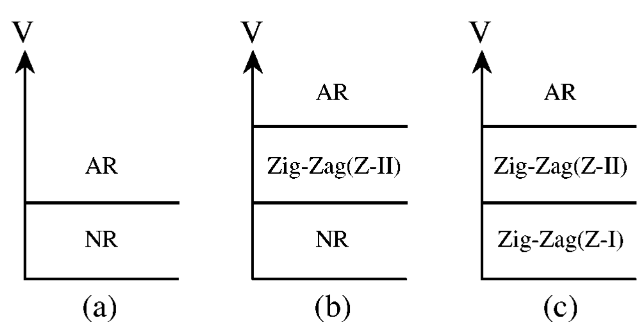

FIG. 5. Typical routes to the *abnormal* roll (AR) increasing voltage V in three different frequency regimes: (a) $f > f_{AR}$, (b) $f_{AR} > f > f_L$, and (c) $f < f_L$. Here $f_{AR} \simeq 1100$ Hz and $f_L \simeq 600$ Hz at $H_x = 1600$ G are the *abnormal* and Lifshitz frequencies, respectively [14].

present destabilizing processes and the soft mode turbulence (SMT) reported in Ref. [22] in the homeotropic system without a magnetic field. SMT was characterized as a direct transition from the rest state to the spatiotemporal chaos via a supercritical bifurcation. In the NR regime it showed a transition from a slow dynamics to a fast one at a characteristic voltage V^* (i.e., $\varepsilon^* \simeq 0.1$) [22]. These aspects of SMT appear to be related to the present instability and destabilizing mechanism. Therefore, the detailed investigation of the instability may give a hint for understanding the characteristics of SMT. In this analogy V^* may correspond to the transition point (V_{NA}) from NRs to ARs [22]. However, in order to prove our speculation, further investigation will be necessary.

Rudroff and Rehberg reported that the restabilization process of ARs beyond the zigzag instability in the planar system was enforced by the lateral boundaries in a long channel parallel to the roll axis [23]. However, as the restabilization of straight rolls (ARs) in the present homeotropic system is observed in sufficiently large aspect ratio ($\Gamma_y = 192$), it must be an intrinsic property of the instability and cannot be considered to be of a similar origin.

III. SUMMARY

In the present study, two types of normal rolls (NRs and ARs) in the homeotropic system in a magnetic field are observed and the instability (the abnormal instability) is reported in the oblique roll regime as well as in the normal roll regime. The instability gives rise to the $y \rightarrow -y$ symmetry breaking on the director. In addition, zigzag patterns, such as two types of zigzag patterns *Z*-I and *Z*-II, have been found in the oblique roll regime. They may occur due to an in-plane distortion by coupling between the director field and the velocity field beyond a new threshold. *Z*-I and *Z*-II in zigzag rolls correspond to NRs and ARs in straight roll patterns, respectively. That is, it can be said that a zigzag pattern of *Z*-I is due to the normal-type zigzag instability, while one of *Z*-II is due to the *abnormal-type zigzag* instability.

ACKNOWLEDGMENTS

This work has been partly supported by a Grant-in-Aid for Scientific Research from the Ministry of Education, Science, Sports and Culture in Japan (Grant Nos. 08454107 and 10440117).

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[13] The arguments here are based on the *small* n_y gradients, i.e., the in-plane distortion of the director **n**. In planar system, however, the boundaries of the domains with oppositely twisted director due to the *large* n_y gradients for large voltages (i.e., ε) are visible with the standard optical setup [9]. In the same way, the boundaries of the domains with the *large* $\pm\alpha$ in our present homeotropic system stated below are also visible with no cross-polarizer set [see Fig. 2(a) herein and Fig. 9 of Ref. [14]]. It is a well-known fact.
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