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Prewavy Pattern: a Director-Modulation Structure in Nematic Liquid Crystals

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A periodic structure (the prewavy pattern) found below the onset of electroconvection in a homeotropically aligned nematic liquid crystal is reported. It is characterized by a periodic modulation of the director in the xy-plane, which can be distinguished from electroconvection patterns with modulation in the xz-plane. The prewavy instability is investigated in detail; a phase diagram in the frequency-voltage plane, the voltage dependence of the azimuthal rotation angle, the voltage and frequency dependence of the wavelength of the prewavy pattern, and the director field in the prewavy pattern are provided. This prewavy pattern always evolves into the so-called chevrons above onset of convection. The wavelength of the chevrons and the orientation of their alternating zig and zag rolls depend on the director structure of the prewavy pattern.

Keywords: electroconvection; homeotropic alignment; Fréedericksz transition; chevron

INTRODUCTION

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Nematic liquid crystals with negative dielectric anisotropy can exhibit a large variety of electroconvection (EC) patterns. EC is realized by applying electric field ($\mathbf{E} = E_z\hat{\mathbf{z}}$) across a nematic slab (typically thickness d = 10 - 100μm) sandwiched between two electrode plates. In general, EC in nematics can be described by the Carr-Helfrich mechanism which allows convection of fluid mass with intermediation of electric charge current driven by an applied ac-voltage[1]. There are two distinct frequency regimes which are separated by the cut-off frequency f_c. In the conduction regime ($f < f_c$) the threshold voltage V_c for the primary instability of EC pattern which appears as normal rolls or oblique rolls has a strong frequency dependence diverging as f approaches f_c, while it has a weak frequency dependence ($V_c(f) \sim f^{1/2}$) in the dielectric regime ($f > f_c$). Moreover, the wavelength λ of the roll pattern in the conduction regime is of order d, which is distinguishable from that of the dielectric roll pattern (λ << d).

In addition to the above-mentioned roll patterns certain substances may form another type of periodic pattern below the threshold of the EC mentioned above. To observe the so-called *prewavy pattern* the sample must be placed in between two crossed polarizers. The pattern manifests then itself as a series of brighter and darker stripes, running along y, perpendicular to the director orientation (along x), similarly to the EC pattern, however, with a wavelength much larger than d ($\lambda \approx 4d$ - $6d$). Very recently, a reinvestigation of the prewavy pattern has been started[2], long after it was first reported in Refs. [3, 4]. The renewed interest in the prewavy pattern is owing to the periodic modulation of the director in the xy-plane in contrast to normal rolls in EC with the modulation in xz-plane. Moreover, it plays a crucial role in the formation of the so-called chevron pattern at higher applied voltages[2]. Though the prewavy pattern was first seen already in the 70s, the mechanism of formation of this pattern has not been explored yet, despite of the fact that in the past decades several theoretical models have been developed to explain the appearance of various patterns induced by electric fields, observed in planar and/or homeotropic nematics.

Up to now it has been reported that the prewavy pattern appears in several cases[2-8]. In this paper we concentrate on the prewavy pattern observed in homeotropic ($\mathbf{n} = (0, 0, 1)$ in the initial state) samples of strongly doped *p*-methoxybenzilidene-*p*'-*n*-butylaniline (MBBA) possessing high electrical conductivity, and present detailed characteristics of the prewavy pattern.

EXPERIMENTAL

In the experiments a nematic liquid crystal MBBA was filled between two parallel glass plates whose surfaces were coated with transparent (ITO) electrodes. The distance $d = 50\mu\mathrm{m}$ between the two electrodes was maintained with a polymer spacer (Mylar), and the lateral size of the electrodes was $1 \times 1\,\mathrm{cm}^2$. In order to achieve a homeotropic alignment, the surface of the glass plates was treated by the surfactant *n*-*n*'-dimethyl-*n*-octadecyl-3-aminopropyl-trimethoxy silyl chloride (DMOAP). The electric conductivities of the sample were $\sigma_{\parallel} = 9.08 \times 10^{-7}\ \Omega^{-1}\mathrm{m}^{-1}$ and $\sigma_{\perp} = 7.37 \times 10^{-7}\ \Omega^{-1}\mathrm{m}^{-1}$, which were obtained by doping MBBA with 0.01 wt% of tetra-*n*-butyl-ammonium bromide (TBAB). The dielectric constants of the sample were $\varepsilon_{\parallel} = 4.33$ and $\varepsilon_{\perp} = 4.62$. The subscripts $\parallel$ and $\perp$ denote the orientations parallel and perpendicular to the director $\mathbf{n}$, respectively.

In all measurements the temperature of the cell located in a Teflon-wrapped copper cavity was stabilized at 30.0±0.1°C by an electronic control system. Applying an alternating electric voltage V (root-mean-square) to the cell the nematic slab was subjected to an electric field $\mathbf{E} = (\sqrt{2}\,V/d)\cos(2\pi ft)\hat{\mathbf{z}}$.

RESULTS AND DISCUSSION

Increasing the applied voltage V above $V_F \approx 3.5$ V the initial homeotropic state undergoes the bend-Fréedericksz transition. After enough time (typically 50 - 60 min) the azimuthal direction of the tilted director becomes uniform, namely the Fréedericksz state becomes homogeneous in the *xy*-plane. We fixed the direction of the *x*-axis of our frame of reference along the projection of the director onto the *xy*-plane (the C-director) in this homogeneous Fréedericksz state.

Figure 1 shows a phase diagram in the frequency-voltage plane. The Fréedericksz threshold voltage V_F was independent of the frequency

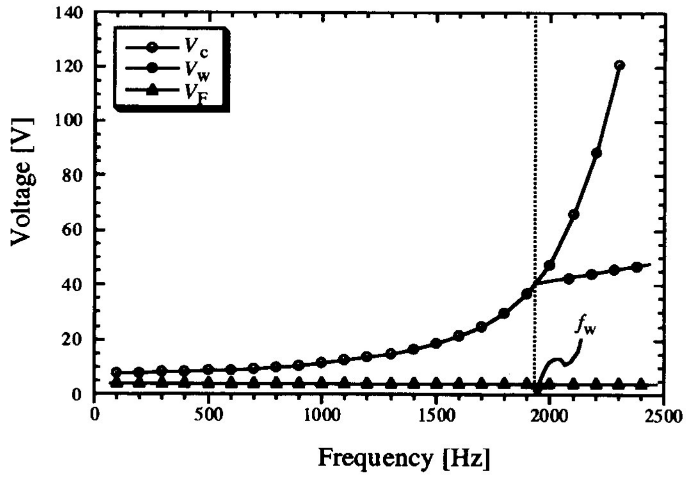

FIGURE 1 Phase diagram of the homeotropic MBBA in the frequency-voltage plane. Here V_F, V_w and V_c indicate the threshold voltages of the Fréedericksz transition, the prewavy pattern and the electroconvection, respectively. The prewavy pattern appears at high frequencies, above a characteristic frequency f_w ($<f_c$), between V_w and V_c.

f in the studied frequency range. In contrast to this the threshold voltage V_c ($> V_F$) for the onset of EC exhibits strong frequency dependence typical for the conductive regime. At high frequencies, above a certain characteristic frequency $f_w \approx 1950$Hz ($<f_c \approx 3000$Hz), on the other hand, there is another threshold line V_w ($\gg V_F$) for a *periodic* pattern. The threshold V_w increases slightly with the frequency ($V_w \propto f$), but no specific change has been detected at (or around) f_c. The prewavy pattern appears at electric voltages much higher than the bend Fréedericksz threshold for the homeotropic system as shown in Fig. 1. At such high voltages the director structure is practically planar everywhere in the sample except a thin boundary layer at each electrodes. Since cells with initial planar orientation can also exhibit prewavy patterns, the orientation in these thin boundary layers do not seem to play an important role in the formation of the prewavy pattern. Hence the homeotropic cell in this voltage range is approximated as a planar cell with unusual (azimuthally degenerate) bound-

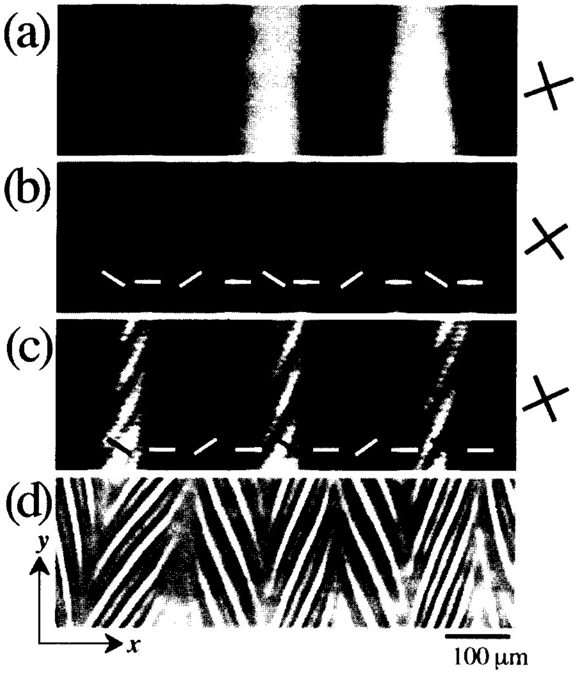

FIGURE 2 Pattern evolution with increasing voltage (at a fixed frequency $f = 2150\text{Hz} > f_w \approx 1950\text{Hz}$). The prewavy patterns at $V = 46.03\text{V}$ (slightly above $V_w = 43.98\text{V}$) (a) and at $V = 67.64\text{V}$ (b) with crossed polarizers. At higher voltage $V = 76.22\text{V}$ the chevron pattern was observed with crossed polarizers (c), and without polarizers (d). The cross at the right-hand side of (a) - (c) indicates the orientation of the crossed polarizers while the bars in (b) and (c) represent the corresponding **C**-director.

ary conditions. Actually the boundary layers allow to avoid the twist deformation along z (which occurs in a common planar cell) and therefore make the observation of the azimuthal modulation much easier.

Figures 2 (a) and (b) show the patterns with increasing voltages V ($> V_w = 43.98\text{V}$) at a fixed frequency $f = 2150\text{Hz}$ ($> f_w \approx 1950\text{Hz}$). The pattern consists of parallel stripes running along y, i.e. they are perpendicular to the **C**-director in the Fréedericksz state (x). Obviously, they have much larger wavelength ($\lambda_w > d$) than that of a normal convection pattern ($\lambda_c \sim d$) in Fig. 2 (d). This pattern often evolves into a sinusoidal wavy pattern with time[2,3]. That is the reason why it is called the *prewavy* pattern. The prewavy pattern has the best contrast when crossed polarizers are used, though it remains visible with parallel polarizer setting. However, removing any of the polarizers makes the pattern almost totally undetectable, similarly to the planar case[4,6,7]. When rotating the crossed polarizers synchronously, the optical intensity of the pattern changes with a 90°-periodicity. At the rotation angle of 45° an optical inversion of the pattern can be detected.

These observations indicate that the prewavy pattern corresponds to a periodic modulation of the in-plane (azimuthal) angle of the director which is symmetric with respect to the initial director orientation in the (homogeneous) Fréedericksz state (see the periodic variation of the C-director in the xy-plane in Fig. 2 (b)). The modulation of the director in the xz-plane, if there is any, must be much smaller than that induced by usual EC patterns.

When the applied voltage is increased above V_c at frequencies below f_w EC appears in the form of the typical normal rolls. In the frequency range of $f_w < f < f_c$, however, an unusual chevron pattern (the defect-free chevron in Ref. [2]) can be observed already at the onset of the EC as shown in Figs. 2 (c) and 2 (d). In this chevron pattern the EC rolls form a nonzero angle with both the x and y axes, hence one always finds alternating zig and zag domains with oppositely tilted roll directions. Consequently there is a double periodicity in the pattern, the smaller wavelength corresponds to that of the EC rolls, the larger one to that of the zig and zag domains.

This chevron pattern must be closely related to the prewavy instability since the EC rolls appearing at V_c are roughly normal to the director in the prewavy pattern just below V_c, as seen in Fig 2 (c). The boundaries between the zig and zag domains correspond to those lines in the prewavy pattern, where the local C-director coincides with the x axis (the initial C-director in the homogeneous Fréedericksz state). Thus the defect-free chevron pattern seems to be a superposition of the prewavy and EC patterns (normal rolls in a system with periodic director modulation).

The voltage dependence of the prewavy pattern has been investigated with crossed polarizers. We defined the in-plane rotation angle α as the angle between the C-director and the x-axis, consequently $\alpha = 0$ in the initial Fréedericksz state. The amplitude α_{max} of the spatially modulated in-plane rotation angle has been measured by rotating the crossed polarizers while the applied voltage was gradually increased in small steps at a fixed frequency $f = 2150$Hz. The threshold $V_w = 43.98$V has been determined as the voltage at which the periodic α modulation first becomes detectable. Slightly above V_w the pattern is still very faint as α_{max} is of order 1° - 5° only. Above another critical voltage $\bar{V}_w = 46.77$V the pattern becomes more intensive, as α_{max} starts to grow steeply with increasing voltage (see Fig. 3). In this voltage range the $\alpha_{max}(V)$ curve may indicate a quasi-pitchfork bifurcation. α_{max} can be fitted by $\alpha_{max}(\varepsilon_w) = \Phi\varepsilon_w^{1/2}$ ($\Phi = 65.3°$) for small ε_w ($\varepsilon_w = (V^2 - \bar{V}_w^2) / \bar{V}_w^2$ is the reduced voltage), but deviates toward a saturation angle of about 45° at higher voltages. The α_{max} observed between V_w and $\bar{V}_w$ may be a kind of premonitory fluctuation which is often observed in convection system[9,10] and laser instability[11]. Crossed polarizers provide high sensitivity for detecting even such

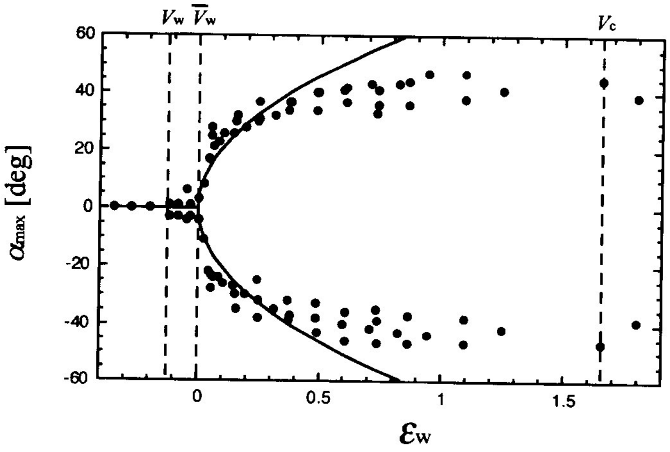

FIGURE 3 The in-plane rotation angle $\alpha_{max}(\varepsilon_w)$ (at $f = 2150$Hz). The prewavy pattern becomes detectable at V_w. The thick lines correspond to a fit of $\alpha_{max}(\varepsilon_w) = \Phi\varepsilon_w^{1/2}$. See the text for details.

a small amplitude of the prewavy pattern.

The prewavy pattern appears and disappears very slowly. Its characteristic time seems to be much slower than that of EC. This manifests also in the slow motion of defects (dislocations) in the prewavy pattern when the system is adjusting the stripe direction and spacing. Therefore, the measurements have been repeated using another method. The applied voltage was first kept at $V_0 = 10$V ($V_F < V_0 < V_w$) until the homogeneous Fréedericksz state was recovered (typically 10 min), and then it was suddenly raised to the desired values and kept there for at least 20 min. This method provided slightly different results. The shape of the α_{max} curve looks very similar to that obtained with the gradually increasing voltages, however, α_{max} has significantly lower values ($\Delta\alpha_{max} \approx 10°$ above $V \approx 50$V) as shown in Fig. 4. An even more striking difference can be observed in the voltage dependence of the wavelength λ_w in Fig. 5. While with gradually increasing voltages λ_w is practically constant until defects appear in higher voltage range, in the case of a sudden raise of the voltage it shows a pronounced voltage dependence, a reduction of λ_w with increasing voltages. The increment of "free energy" from the homogeneous

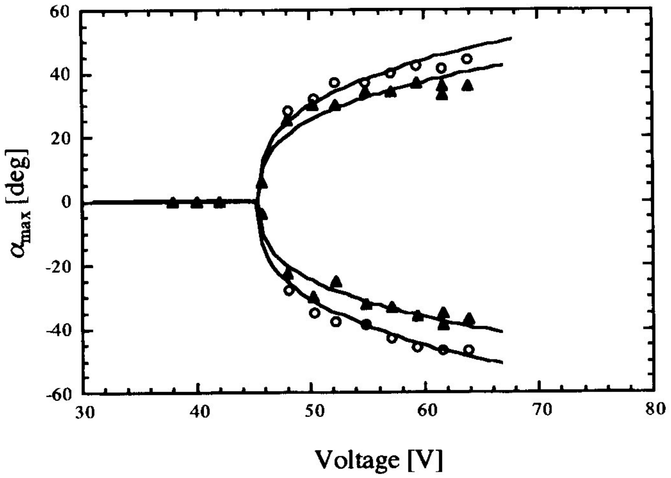

FIGURE 4 The in-plane rotation angle $\alpha_{max}(V)$ measured by gradually increasing (open circles) and suddenly raised (solid triangles) voltages (at f = 2150Hz).

Fréedericksz state to the prewavy state due to the C modulation is larger for larger α_{max} and smaller λ_w. Since the variation of λ_w needs a global change of the system, it is easier for the system to adjust α_{max} rather than λ_w when the external field is changed. Therefore for gradually increasing voltages λ_w remains constant and instead α_{max} becomes larger than for suddenly increasing voltages.

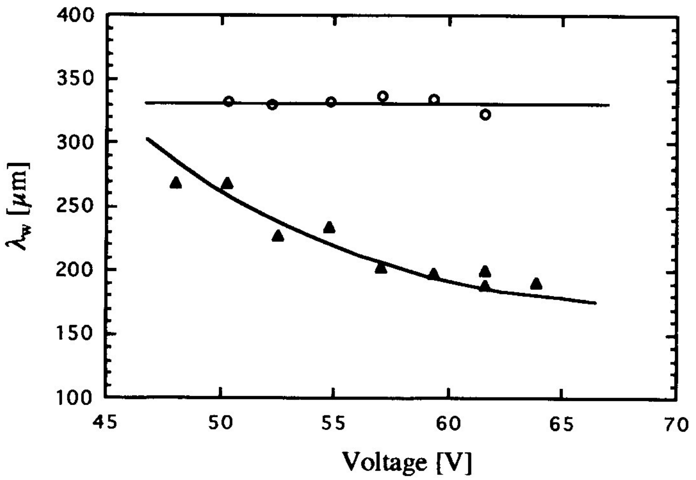

FIGURE 5 The wavelength λ_w of the prewavy pattern measured by gradually increasing (open circles) and suddenly raised (solid triangles) voltages (at $f = 2150$Hz). The solid lines are to guide the reader's eye only.

The frequency dependence of the characteristics of the prewavy pattern has been checked too. The wavelength λ_w measured at a fixed control parameter ε_w seemed to remain constant in the frequency range of $f_w < f < 5$kHz. Moreover, no pronounced frequency dependence of the $\alpha_{max}(\varepsilon_w)$ curve was found ($\Phi \approx$ const.).

Besides visual observation of the prewavy pattern the light intensity transmitted by the cell between crossed polarizers was also monitored by a fast photodetector attached to the polarizing microscope. No modulation with the frequency of the applied field or its upper harmonics could be detected in the transmitted intensity. It proves that the C-director does not oscillate with the applied field.

In order to decide whether there is any flow associated with the pattern, small polystyrene spheres (Micropearl of Sekisui Chemical Co., Ltd.) of 3.88 μm diameter were introduced into the cell. Motion of the particles could be followed by the microscope in the *xy*-plane as well as in the *z* direction (particles went out of focus of the microscope in the latter case) until they were trapped by defects at the surfaces. In the normal EC rolls ($V > V_c, f < f_w$) trajectories of the particles formed closed loops in the *xz* plane, indicating a convective motion as expected. In the defect-free chevrons ($V > V_c, f > f_w$) similar trajectories could be observed, only the plane of convection was rotated to be perpendicular to the EC rolls in the chevrons. In the prewavy pattern, however, a flow along *z* could not be detected. Instead, the particles often show two typical kinds of flow. In one case they were moving parallel to the stripes ($v_x = 0$, $v_y \neq 0$), typically along the lines with $\alpha = 0$. The direction of the motion seemed to be opposite in the neighbouring $\alpha = 0$ lines, the reversal of flow direction occurred at dislocations (the dead end of the stripes) or at cell boundaries. These observation indicate that in a defect-free prewavy pattern the flow velocity has a v_y component only which is spatially periodic. In the other case the particles were moving almost normal to the stripes, having only a small v_y component ($v_x >> v_y$). Since the sign of the v_y component was opposite in neighbouring white lines, their overall motion seemed to be zig-zag. These motions may indicate that the flow correlates with the prewavy pattern. However, one cannot conclude that the prewavy pattern is induced directly by flow, because the flow relaxed much faster than the prewavy pattern when the applied voltage was suddenly decreased below V_w.

CONCLUSIONS

We report the prewavy pattern in homeotropic MBBA with high electric conductivity. It is characterized by a periodic modulation of the director in the *xy*-plane and is found below the onset of electroconvection. The defect-free chevron appears as a superposition of normal rolls of EC on the prewavy pattern. Therefore the wavelength of the chevron and the orientation of its alternating zig and zag rolls depend on the director structure of the prewavy pattern. The prewavy instability is experimentally investigated and discussed in detail; a phase diagram in the frequency-voltage plane, the voltage dependence on the azimuthal rotation angle, the voltage and frequency dependence on the wavelength of the prewavy pattern, and the director field in the prewavy pattern are provided. Unfortunately, however, we cannot reach crucial understanding of the formation mechanism for the prewavy pattern yet.

In the renewed research, the prewavy pattern interests us as a background of the defect-free chevron in doped MBBA[2], where since the dielectric threshold V_c is much larger than V_w, the dielectric pattern cannot be observed due to experimental risk. In thinner cell with lower electric conductivity, however, the transition from prewavy to dielectric chevrons with increasing voltage can be observed. That is a direct proof for the existence of a mechanism other than the Carr-Helfrich one in doped MBBA. Moreover patterns looking like the present prewavy in homeotropic MBBA could be observed also in other substances, e.g. in the thick homeotropic doped Phase 5A. It means that prewavy is not a privilege of MBBA.

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