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Domain instabilities in the planar texture of smectic A liquid crystals

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Domain instabilities have been observed in the planar texture of smectic A liquid crystals. The influence of the material parameters and of the applied field on the characteristics of this phenomena have been studied.

In recent years some electro-optic effects [2–8] with immediate practical value have been found and have been investigated in smectic A liquid crystals with their characteristic layered structure [1] in the molecular alignment. For investigations relating both to a smectic A with positive dielectric anisotropy ($\Delta \varepsilon = \varepsilon_{\parallel} - \varepsilon_{\perp} > 0$) [2–6] and for a smectic A with $\Delta \varepsilon < 0$ only the homeotropic texture behaviour has been studied with an electric field [2, 7]. However, the type of electro-optic effects occurring in liquid crystals depend strongly on the initial molecular orientation. In this paper we report the results of a smectic A planar texture behaviour subject to an electric field; these reveal new instabilities as domain images in this phase.

This effect was discovered in the liquid crystal with a structural formula

$$H_{21}C_{10}O - OC_4H_9$$
 (A)

and with transition temperatures:

 $C \xleftarrow{34^{\circ}C} S_B \xleftarrow{54^{\circ}C} S_c \xleftarrow{59^{\circ}C} S_A \xleftarrow{83^{\circ}C} N \xleftarrow{93^{\circ}C} J$

This effect has been observed in other liquid crystals having a smectic A phase with negative dielectric anisotropy. In particular, the liquid crystal

 $H_{13}C_{6}O - OC_{6}H_{13}$ (B)

has been investigated.

It turns out that the phenomenon observed is found for all smectics with $\Delta \varepsilon < 0$. But a relatively low threshold voltage and good contrast have been found for compound A; therefore, quantitative data are reported for this substance.

The device was placed on the stage of a polarizing microscope, which allowed us simultaneously to carry out electro-optic measurements and to observe texture transformations. The thickness and the effective area of the electro-optic cell were $20 \,\mu m$ and $0.8 \,\mathrm{cm}^2$, respectively. The homeotropic texture was formed, spontaneously, on cooling the sample from the nematic phase. The qualitatively planar texture used in our studies was obtained by the homeotropic-planar transition produced by a high-frequency electric field [7].

When applying a low frequency electric field to the prepared cell up to a value $U_{th}^{(0)}$, visible changes do not take place in the sample texture. $U_{th}^{(0)}$ is the threshold voltage needed to form a 'low frequency instability', discovered earlier [8]. Note that this instability does not cause changes of the sample's optical properties. This was demonstrated by the motion of small foreign particles perpendicular to the optic axis.

At the relatively high voltage of $U_{th}^{(1)}$ we observed an instability as domain images with a period of order of the cell thickness $l_1 \approx d$ (see figure 1 (*a*)). Until the voltage of $U_{th}^{(2)}$ is reached this domain period is constant. At $U_{th}^{(2)}$ the domain image is reformed. The primary domains are substituted for the domain with a large period $l_2 \approx 2d$ and with greater contrast (see figure 1 (*b*)).

Both the primary and secondary domains were formed along the optic axis of the mesophase. For this reason, when the polarization vector of the incident ray is perpendicular to the optic axis, there is maximum contrast. After switching off the voltage the domain image disappears and the initial planar texture is reestablished with a relaxation time of 50–100 ms.

Due to the domain formation the intensity of light passing through the liquid crystal is modulated and the modulation frequency is doubled (figure 1(c), (d)). The modulation depth

$$M = \frac{\tau_{\max} - \tau_{\min}}{\tau_{\max} + \tau_{\min}}$$

increases with increasing U for voltages changing within the range $U_{th}^{(1)} \leq U \leq U_{th}^{(2)}$. For $U > U_{th}^{(2)} M$ decreases slightly (see figure 2). With decreasing temperature the threshold voltage of domain formation increases. As shown in figure 3 the temperature dependence of $U_{th}^{(2)}$ is stronger than $U_{th}^{(1)}$.

The domains were not formed by application of a d.c. electric field to the sample, even when applied up to the breakdown voltage. Domain instability is observed only under a low frequency electric field within the narrow frequency range $20 \le f \le 100 \text{ Hz}$ (see the insert in figure 3).

Further work has been directed towards the elucidation of the influence of changes in the material on the domain instabilities characteristics, in order to understand the mechanism of domain formation. It is found that by doping the sample with conductive additives (for example, tetrabutylammonium bromide (TBAB)) the frequency range of the domain existence could be extended. For example, if for σ_{sp} is $2 \cdot 1 \times 10^{-10} \Omega^{-1} \text{ cm}^{-1}$ the domains are observed until f_0 is 90 Hz; then for σ_{sp} of $5 \cdot 1 \times 10^{-9} \Omega^{-1} \text{ cm}^{-1}$, f_0 equals 500 Hz. For samples doped in this manner $U_{th}^{1,2}$ were found to vary as $f^{1/2}$. Doping the liquid crystal with TBAB increases both its average value of electroconductivity and anisotropy. $\sigma_{\perp}/\sigma_{\parallel}$. It turns out that in all cases the threshold voltage is correlated with $\sigma_{\perp}/\sigma_{\parallel}$, but not $\bar{\sigma}_{sp}$ (see figure 4). As shown in figure 4, the low frequency instability threshold of $U_{th}^{(0)}$ does not depend on the anisotropy. This is in agreement with the conclusion reported for the isotropic mechanism of this instability formation [8].

In order to distinguish the role of the electroconductivity anisotropy the liquid crystal A was doped with the nematic additive

The characteristic feature of this additive is that in small concentrations (C ≤ 5 per cent) it decreases the smectic A phase electroconductivity anisotropy. But σ_{sp} and other parameters of the mixture are not changed. As shown in figure 5 there



Figure 1. Photographs of (a) primary and (b) secondary domains and oscillograms of the signal, (c), (d) modulated by corresponding domain structure. Above it is displayed the a.c. electric field applied to the cell (f = 20 Hz, $V_c = 200$ V, $V_d = 275$ V).



Figure 2. The dependence of the modulation depth of light passing through primary and secondary domain on the voltage of the applied field.



Figure 3. Threshold voltage temperature dependences for the (1) primary and (2) secondary domains, measured at f = 20 Hz. In the insert the frequency dependences of the threshold voltages at $t = 82^{\circ}$ C are shown.

a good correlation between $U_{\rm th}^{(1)}$, $U_{\rm th}^{(2)}$ and $\sigma_{\perp}/\sigma_{\parallel}$, i.e. decreasing the electroconductivity anisotropy leads to increasing threshold voltages.

Mixing the liquid crystals A and B causes the value of the dielectric anisotropy $(\Delta \varepsilon = \varepsilon_{\parallel} - \varepsilon_{\perp})$ to change within the range $-0.5 \le \Delta \varepsilon \le 0.5$. The investigation of similar mixtures has indicated that $\Delta \varepsilon$ does not influence to any significant extent the threshold voltage value.



Figure 4. Threshold voltage concentration dependences of (1) the low frequency instability, (2) the primary and (3) secondary domains and (4) of the electroconductivity anisotropy measured at different temperatures, where TBAB is used as a dopant: (a) $t = 80^{\circ}$ C; (b) 73°C; (c) 64°C.

As we have seen, the liquid crystal A is a convenient substance with which to observe the instability. For this reason a study of the properties of this substance is important. As shown in figure 6, this compound is not distinguished from other liquid crystals by its σ and $\Delta \varepsilon$ values. Its unusual feature is a large value of the electroconductivity anisotropy, which varies over the range $4 \le \sigma_{\perp}/\sigma_{\parallel} \le 7$. Indeed, when this substance is doped the anisotropy $\sigma_{\perp}/\sigma_{\parallel}$ can reach 10.

In conclusion, to account for the experimental facts we can make the following inferences concerning the possible mechanism of domain formation.

A low frequency instability occurs when a low frequency electric field of small amplitude is applied to the smectic A planar texture with $\Delta \sigma < 0$ and $\Delta \varepsilon < 0$. Its formation is explained by the so-called isotropic mechanism [9], according to which the non-uniform distribution of the space charge over the cell thickness is taken into account. The electric field interaction with this space charge leads to the rotational



Figure 5. Threshold voltage concentration dependences (1) of the low frequency instability (2) of the primary and (3) secondary domains and (4) of the electroconductivity anisotropy, measured at 80°C. A nematic additive is used to dope the sample.



Figure 6. Temperature dependences of the electroconductivity components σ_{\parallel} and σ_{\perp} (1,3) and of the electroconductivity anisotropy of $\sigma_{\perp}/\sigma_{\parallel}$ as well as the dielectric anisotropy (4).

flow of the medium near the electrode regions on the thickness of the Debye distance, $L_{\rm D}$. It is confirmed by the circular motion perpendicular to the optic axis of foreign particles specially doped into the liquid crystal. The rotation speed increases with voltage and the whirls penetrate deeply into the bulk liquid crystal. The resultant flow destabilizes the layer's equilibrium state and leads to its undulating modulation. The

wave vector of this layer modulation is perpendicular to the smectic optic axis. The moment, caused by the electroconductivity anisotropy $\sim (\sigma_{\perp}/\sigma_{\parallel})/E^2$, increases the distortion. When linear polarized light with its wave vector perpendicular to the optic axis is passed through such a modulated structure, the optical pattern observed in crossed polarizers appears as alternating dark and bright stripped domains, directed perpendicular to the layer plane, i.e. along the optic axis.

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