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Observations of flexo-dielectric walls in a bent-core-calamitic nematic liquid crystal♣

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Experimental observations on the electrooptic behaviour of the bent-core-calamitic 4-((3-(4-(4-(decyloxy)benzoyloxy)benzoyloxy)phenylimino)methyl)-3-hydroxyphenyl 4-(6-(4'-cyanobiphenyl-4-yloxy) hexyloxy) benzoate in the nematic phase with a positive dielectric anisotropy were performed for the first time. The efforts were concentrated mainly on the flexoelectric behaviour of the nematic for the case of a d.c. or very low-frequency (up to 0.05 Hz) voltage applied across a highly tilted or homeotropic nematic layer with a thickness of 15 μm. Various flexo-dielectric domains, such as cross-like domains, π-walls, longitudinal flexo-dielectric walls, etc. were observed for voltages below 8 V. The cross-like domains were observed in initially homeotropic nematic regions. The π-walls were observed in highly tilted nematic regions. The cross-like domains and the π-walls can be erased at the threshold by applying an additional high-frequency (5 kHz) voltage of 1-2 V_{rms}. Our interpretation is that all these flexoelectric domains arise from the inhomogeneity of the electric field created by injection of ions from one of the electrodes.

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1. Introduction

According to [1, 2] the first synthesis of banana-shaped liquid crystals happened in 1980. References concerning banana-shaped mesogens having both nematic only or nematic and smectic phases, synthesized in the period 1993-2002 can be found in the papers published by Pelzl et al. [3] and Jakli et al. [4]. In recent years, there has been increasing interest in the flexoelectricity in the banana-shaped nematics [4, 5]. The experiments showed a much stronger flexoelectricity in these. A few years ago, a low-molar-mass, monodispersive, bent-rode dimer exhibiting biaxial nematic and smectic-A phases was synthesized and studied by Yelamaggad et al. [6] (see Fig. 1).

2. Materials, sample preparation and set-up

During our experiment, we have studied the same liquid crystal, synthesized by Hiremath and Yelemaggad. This liquid crystal is monotropic: upon heating, it has only a nematic phase and upon cooling it has additionally two smectic phases [6]. We studied it under heating only. Initially, this liquid crystal had the following phases and phase transitional temperatures:

$$\text{Cr-}137.8^0\text{C-N-}160^0\text{C-I} \quad (1)$$

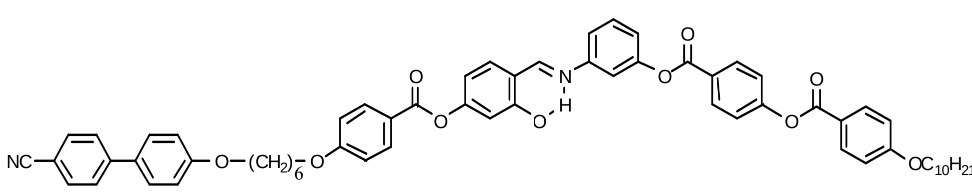

Fig. 1. Structural formula of a bent-core-calamitic compound.

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The liquid crystal samples were prepared with the aid of two ITO-coated glass plates separated by Teflon sheets (spacers), 15 μm thick. Technical problems did not allow us to use thinner cells. The liquid crystal cell was put in a Mettler hot stage, type FP-82. The temperature was varied from room temperature up to 200^0C by a home made electronic regulator. The sample temperature was measured by a high-precision digital thermometer. It was confirmed that the nematic-isotropic phase transition temperature under heating stays near to the original data (159 ^{0}C, Eq.1). In order to observe the electrooptical behaviour of this new type of nematic, we used a d.c. voltage up to 15 V or a very low-frequency voltage with a frequency up to 0.05 Hz. Additionally, we applied a high-frequency erasing voltage of 5 kHz and amplitude from 1 to 10 V_{rms}. Sometimes, the d.c. and a.c. voltages were applied simultaneously. The d.c. current was measured by a high-precision digital multimeter, type Keithley 179. While the experiment was developing, we recorded some decrease in the phase transitional temperatures, which at the end of the experiment had the following values:

$$\text{Cr-131}^0\text{C-N-150-}^0\text{C-I} \qquad (2)$$

Moreover, we noted that these phase transition temperatures could decrease further with several degrees of ^{0}C, when the cell was under the influence of a d.c. voltage above 2 V for a long time (at least several hours). Metal foil clips, usually giving mechanical stability to the cells were not used, due to the specificity of the Mettler hot chamber and the high temperatures at which the nematic phase exists. Instead, we employed a special two-side sticking band which cannot be destroyed at as high a temperature as 200^0C. The resistance of the prepared cells was above 20 MΩ (for a liquid crystal film area of c.a. 3 sq. mm). The dielectric anisotropy $\Delta\varepsilon$ was measured to be + 5.9 ($\varepsilon_{//}$ = 11.7, $\varepsilon_{\perp}$ = 5.8) at T = 135 ^{0}C [7]. The other material parameters such as elastic constants, viscosity coefficients, flexoelectric coefficients, etc. have not been examined.

In our experiment, ITO coated glass plates were rubbed by hand, to ensure a high-tilted orientation of the nematic. The presence of a cyano group at one end of the molecule (see Figure 1) facilitates the development of homeotropic nematic anchoring at the ITO electrode surface. Indeed, during the experiment, we observed a homeotropic orientation of the nematic, with several imperfections in the view of the microscope. Additionally, the transient gradient in the temperature was found to produce a high-tilted nematic. After the stabilization of the temperature, however, we again observed a homeotropic orientation of the nematic. Also, we noticed a nearly-planar nematic layer with a Schlieren texture approaching closely (fractions of a ^{0}C) the nematic-isotropic phase transition. In our opinion, in this case the glass plates were covered by a thin isotropic layer.

3. Electro-optical observations

We have observed cross-like domains and π-walls [8] in homeotropic and high-tilted nematic layers excitated by a d.c. voltage with amplitudes between 2.5 – 6 V. A typical picture is shown in Fig. 2, for a voltage of 5 V. It was observed that the domains are formed near the anode. They can be easily viewed when the (+) pole is applied to the upper electrode, from the side of the observer (all observations and photos were done with a polarizing microscope, type MPI 5 by PZO, Warszawa, Poland). The domains appeared at a d.c. voltage of 2.5 V – 3 V, depending on the value of the temperature (see Table 1). They could be erased with a low, 5 kHz a.c. voltage (see Table 1, showing the voltage for complete erasing of the domain pattern). Increasing the applied d.c. voltage to 5 V increased the static deformations inside the domains, and in some cases led to their slight contractions and simultaneously caused an increase in the homeotropic regions. These higher voltage effects distinguish the cross-like domains observed in the nematics with positive dielectric anisotropy from those observed in negative ones [8]. A further rise of the d.c. voltage to 7 V led to the appearance of complex electro-hydrodynamics, which destroyed the domains shown in Fig. 2 (see Fig. 3), in contrast to the calamitic nematics with positive dielectric anisotropy in which the injection electro-hydrodynamic domains were cross-like or joined several cross-like domains.

The additional application of an a.c. voltage with a frequency f = 5 kHz and an amplitude of 5 V_{rms} completely removed the electro-hydrodynamics, and the static domains and orients the nematic homeotropically (the changes started at an a.c. voltage of 3 V_{rms}).

4. Discussion and conclusions

In usual calamitic nematics with positive dielectric anisotropy, similar domains have been observed by many authors, and some of them have been explained by an electro-hydrodynamic injection mechanism [8, 9 and citations therein]. Other authors explain such domains taking into account the flexoelectric mechanism [10,11] or the mechanism of surface polarization [12]. In our opinion, the observed domains are flexoelectric ones (more correctly they are flexo-dielectric walls [8]) coupled with the gradient of the electric field due to unipolar injection from the anode, forming a

Fig. 2. Cross-like domains and π-walls above the threshold (U = 2.5 V) viewed under a polarizing microscope in crossed nicols. The applied d.c. voltage is 5 V, the (+) pole is on the upper electrode surface coinciding with the microphotograph focal plane, T = 142 ^{0}C. The long side of the photograph measures 790 μm.

Table 1. The threshold and erasing voltages as a function of temperature.

U_{dc} [V]	U_{ac} [V_{rms}]	T [ºC]
3.0	1.1	131.8
3.0	1.1	135.7
2.9	1.1	139.6
2.5	1.8	146.5

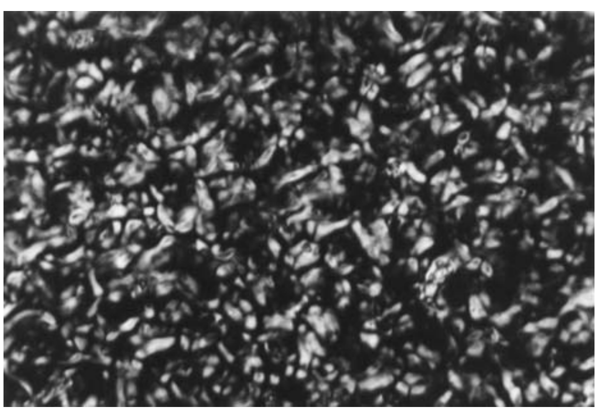

Fig. 3 Electro-hydrodynamics at a 7 V d.c. voltage, destroying the cross-like domains and π-walls viewed under a polarizing microscope in crossed nicols. The (+) pole is on the upper electrode, T = 142 ^{0}C. The long side of the photographmeasures790 μm.

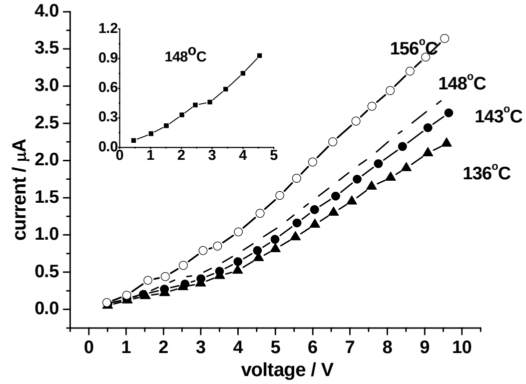

Fig. 4. Current-voltage characteristics of the nematic layer at: 136°C, 143 °C, 148 °C. The upper curve is drawn at 156 ^{0}C, in the isotropic phase.

space charge, reducing the field near it. This is confirmed by the nonlinear current-voltage characteristics shown in Fig. 4 (the appearance of a slope reduction region between 2 and 3 V - see the inset graph).

It is evident that the injection is weak, but sufficient to create various kinds of flexo-dielectric walls and cross-like domains. In addition, the surface orientation of the nematic is of crucial importance for this domain type. Furthermore, the prolonged application (hours) of a d.c. voltage as low as 3 V can change the form of the domains and can transform the cross-like domains into parallel walls with a smaller period compared to the size of the cross-like domains.

In conclusion, we recorded flexo-dielectric walls in a new type of banana-shaped-calamitic nematic layer, due to the gradient of the electric field. The observations confirm the universality of the gradient flexoeffect discovered by the Sofia LC Group [13], for the new case of banana-like nematics.

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