Enhancement of Freedericksz's threshold voltage of nematic liquid crystals in thin cells

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We report the measurement of Freedericksz's threshold voltage (V_{th}) in two pure calamatic nematic as well as a mixture of a calamatic nematic and a bent core (banana) liquid crystal at different thicknesses, in the ranges 1–3 μm and 7 to 15 μm cells. Rubbed polyimide-coated glass plates are used to get planar alignment of the nematic director. We show that the transmitted intensity varies gradually across the transition region with the applied voltage in the thin cells compared to the thick cells. We find a significant enhancement of V_{th} in thin cells in both the compounds and in the mixture and the enhancement is larger at as the temperature is lowered in the nematic phase. Part of it is contributed by the enhancement of the orientational order parameter ($V_{th}^2 \sim S$) in thin cells. Finally we discuss the role of ionic contribution to increase the space charge polarization in thin cells in further enhancing the threshold voltage.

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1. Introduction

Liquid crystals are orientationally ordered liquids, made of aniso-tropic shape [1] organic molecules. There are a variety of liquid crystall; ine phases − among them nematic is the widely-used one. The average orientation direction of the molecules is specified by a unit vector $\hat{n}$ called the director which is *apolar* in nature in the nematic (N) phase.

The material combines anisotropies in dielectric, optical, and other properties with a facility to flow and hence exhibits remarkable electro-optic effects, which are exploited in flat panel liquid crystal display (LCD) devices [2]. When the liquid crystal is confined between the substrates in the presence of electric or magnetic fields the competition between the effects of wall and the effects of external field results in a director distribution beyond a particular field. The reorienting torque arises through a coupling between an external field and the correspond-ing susceptibility anisotropy. Twisted nematic display devices make use of electric field (E) effects in materials with positive dielectric anisotropy ($\Delta\varepsilon = \varepsilon_{||} - \varepsilon_{\perp} > 0$), which favors an alignment of the director parallel to the electric field. The response time for nematic is $\tau \sim \eta d^2 / \pi^2 k$ [1] where η is the effective viscosity, K is an effective curvature elastic constant and d is the sample thickness. A typical LCD cell has a thickness ~5 μm hence a low response time. The response time of the device can be reduced considerably by reducing the sample thickness. In addition, in the bistable twisted nematic displays [3,4] and surface-stabilized ferro-electric devices [5], the thickness has to be quite small, 2 μm or lower. When the thickness of the sample is reduced, the physical as well as the electro-optical properties of the cell becomes significantly different. The

effect of cell thickness on the various physical properties of nematic liquid crystal was reported by several authors [6–9]. Our recent experiments show that the orientational order parameter of nematic liquid crystals in thin cells is enhanced considerably with respect to that in thick cells in all the compounds with aromatic cores [6]. We have also reported the dielectric properties of nematic liquid crystal in the low frequency range in thin cells [7]. The time dependence of switching time in a thin boundary layer bistable nematic display has been discussed by Thruston et al [8]. Mora et al have reported the Freedericksz's transition measurements on the anisotropic polymer-stabilized liquid crystal network and the dependence of threshold voltage on the thickness was explained due to the coupling of the polymer network and nematic elasticity [9]. However, there are no direct measurements of Free-dericksz's threshold voltage in thin cells (1–3 μm) in the planar-aligned nematic liquid crystals.

In the present paper we report the measurement of Freedericksz's threshold voltage as a function of temperature in two calamatic nematic liquid crystals as well as a mixture of a calamatic nematic and banana liquid crystal in different cell thicknesses and show the effect of space charge polarization on the threshold voltage in thin cells.

2. Experimental

Our main aim was to investigate the effect of cell thickness on the Freedericksz's threshold voltage in nematic liquid crystals. We have chosen two calamatic compounds and a bent core compound. The chemical structures and phase transition temperatures of the compounds are shown in Fig. 1. The compound-1, (PCH-5) obtained from Merck, is highly polar. It exhibits a nematic phase at room temperature and is widely used in display applications. The direction of the permanent dipole moment in this molecule is along the long

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Compound-1, (PCH-5): Cr 30 N 55.5 I

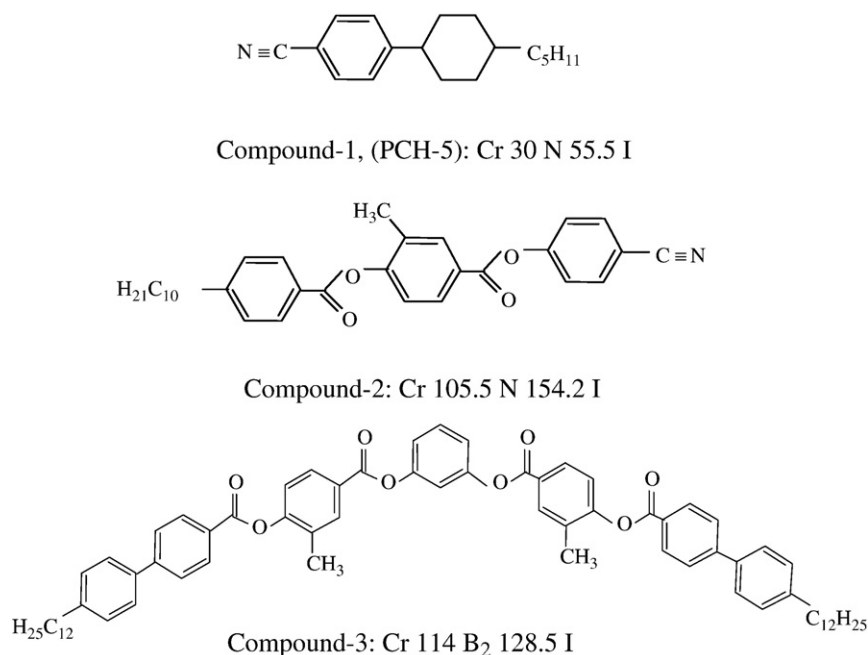Compound-2: Cr 105.5 N 154.2 I

Compound-3: Cr 114 B$_2$ 128.5 I

Fig. 1. Chemical structures, phase sequences and the transition temperatures of the compounds used in the experiments. The transition temperatures are given in degree Celsius.

axis and hence the dielectric anisotropy is positive ($\Delta\varepsilon>0$). The dielectric anisotropy of the nematic is chosen positive so that nematic shows the realignment of director above the Freedericksz's threshold voltage. The compound-2 and compound-3 were synthesized in our laboratory. The compound-2 is also highly polar with a long alkyl chain. It exhibits the nematic phase at relatively high temperatures. The direction of the permanent dipole moment in this molecule is also along the long axis and hence the dielectric anisotropy is positive. The compound-3 is made of bent shaped molecules with a transverse dipole moment and is highly biaxial. The mixture with 50:50 wt.% of the compounds-2 and compound-3, gives rise to a uniaxial nematic of 15-degree temperature range and positive dielectric anisotropy. This mixture was studied to explore the effect of molecular structure on the threshold voltage.

The cells are constructed using ITO coated glass plates. The plates are treated with polyimide and cured at a temperature of 280 °C for 90 min. After curing, the plates are rubbed for homogeneous alignment of the molecules in the nematic phase. The plates were assembled such that the rubbing directions are antiparallel. Glass beads are mixed with epoxy glue, which is spread outside the electrode region to get the required cell thickness (>2 µm). However to make cells of thickness of <2 µm we do not use any spacers. In this case several cells were prepared at a time and a few cells with thickness of <2µm were only selected. The glue used does not affect the liquid crystals and is usually used for making liquid crystal displays. The thickness of the cell is measured at several positions inside the overlapped electrode area using an interferometric technique. The cells with uniform thickness are selected for the experiment. The cell thickness d is found to be constant within 1%. The cell is filled with the sample in the isotropic phase and cooled slowly to the nematic phase, resulting to a good homogenious alignment of the director. The alignment was checked using a polarizing microscope (Leitz ORTHOPLAN). The experimental setup is shown in Fig. 2. The temperature of the sample is controlled by using a heating stage (INSTEC HS1) within an accuracy of 0.1 °C. A helium-neon laser (ORIEL 3 mW, λ = 632.8 nm) beam is used to illuminate the sample. A beam splitter and a reference photodiode (PD2) are arranged to monitor the
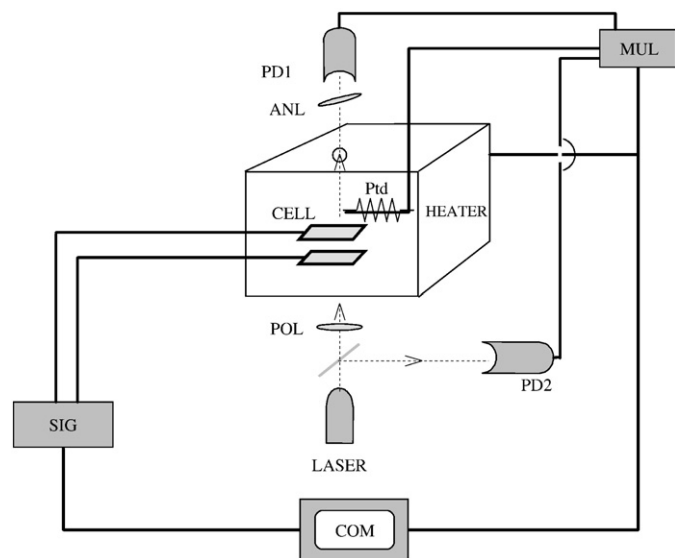

Fig. 2. Schematic diagram of the experimental setup. Photodiodes (PD1, PD2). Polariser (POL), Analyzer (ANL), Multimeter (MUL), Computer (COM). Signal generator (SIG).
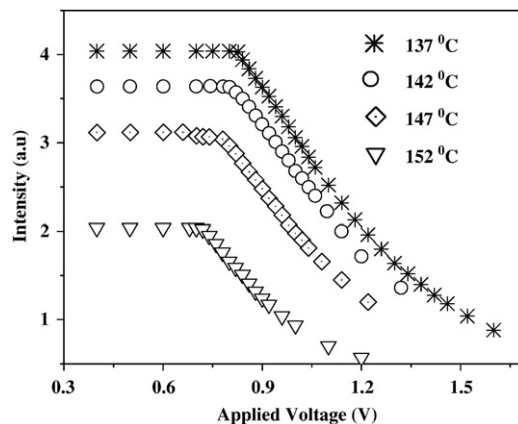

Fig. 3. Variation of transmitted intensity as function of applied voltage of compounds-2 at various temperatures. Frequency: 3037 Hz. Cell thickness: 1.5 µm.
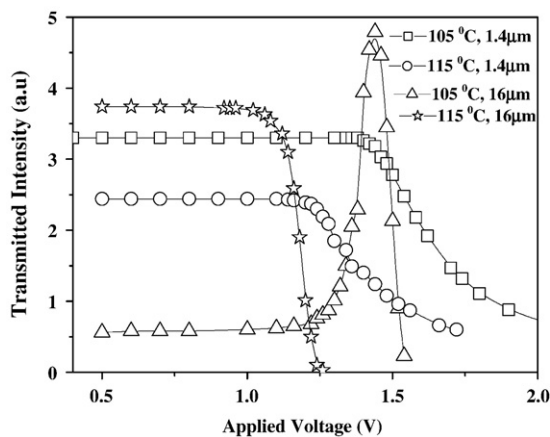
Fig. 4. Variation of transmitted intensity as a function of applied voltage of 50 wt. %:50 wt.% mixture. Open squares and open circles correspond to the data in 1.4 µm cell thickness at temperatures 105 °C and 115 °C respectively. Open triangles and open stars correspond to the data in 16 µm cell at temperatures 105 °C and 115 °C respectively.

Fig. 6. V_{th}^2 at a few temperatures of the compound-2. The open circles and open squares correspond to 1.4 µm and 7 µm thick cells respectively. Dotted lines are drawn as guides to the eye. Frequency: 3037 Hz.

stability of the reference beam intensity. The laser beam is adjusted to be incident on the sample through a polariser and the transmitted intensity is passed through an analyser, which is crossed with respect to the polariser. The optic axis of the sample is set 45° with respect to the polarizer to optimize the measurement. The transmitted intensity is collected by the photodiode PD1. All the photodiodes are connected to a digital multimeter (Keithley-2000). A signal generator (WaveTek) is used to apply an electric field to the cell. The transmitted intensity is measured as a function of applied sinusoidal voltage at various temperatures. The frequency used in the experiment is 3.037 kHz which is typically in the range of LCD application. Finally all the instruments are controlled by a computer (COM). A suitable program is used to run the experiment.

3. Results and discussion

A typical variation of transmitted intensity in 1.5 µm cell as a function of applied voltage at different temperatures for the compound-1 is shown in Fig. 3. The threshold voltage decreases with increasing temperature and the transmitted intensity varies gradually across the transition region. In Fig. 4 we show the variation of intensity in two different cell thicknesses namely 1.4 µm and 16 µm at two different temperatures for the 50 wt.%:50 wt.% of compound-2 and compound-3. The nature of variation of field dependent transmitted intensity in the mixtures at two temperatures is very different. The transmitted intensity in nematic between the two crossed polarisers can be written
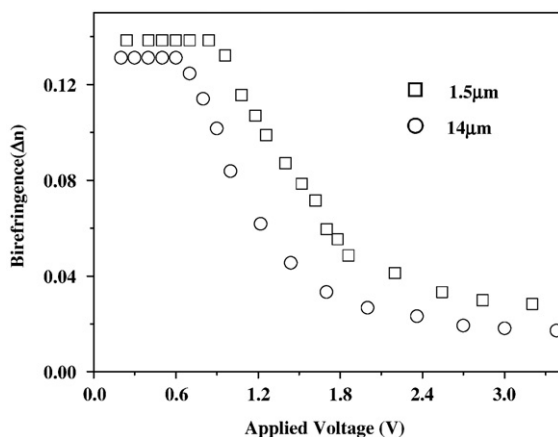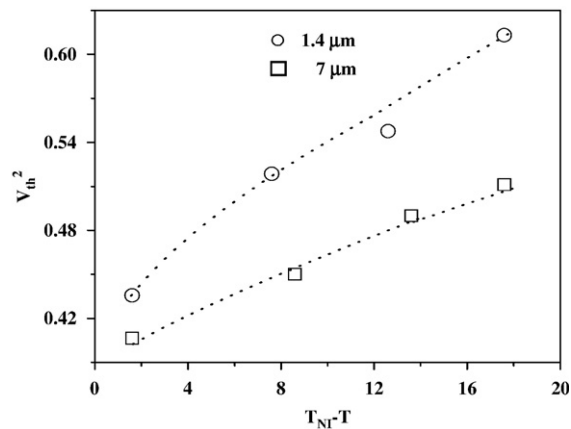
as $I_t = I_o \sin^2(\pi \Delta n d / \lambda)$, where Δn, the birefringence, d, the sample thickness and λ, the wave length of light. The very different nature of the intensity in thick cells arises due to comparatively larger value of the optical path difference ($\Delta n d$). In the mixture the threshold voltage in 1.4 µm is also higher with respect to 16-µm cell at corresponding
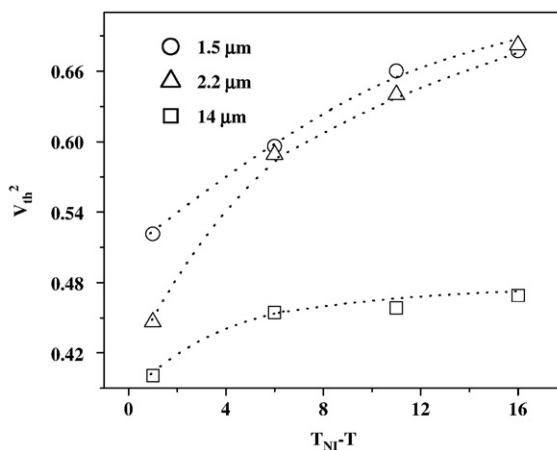

Fig. 7. Variation of V_{th}^2 at a few temperatures in three different cells thicknesses of the compound-3. Open circles (1.5µm), open triangles (2.2µm), open squares (14µm),. Dotted lines are drawn as guides to the eye. Frequency: 3037Hz.

Fig. 5. Variation of birefringence as a function of applied voltage at two different cell thicknesses: 1.5 µm, and 14 µm. Temperature 137 °C. Frequency: 3037 Hz.
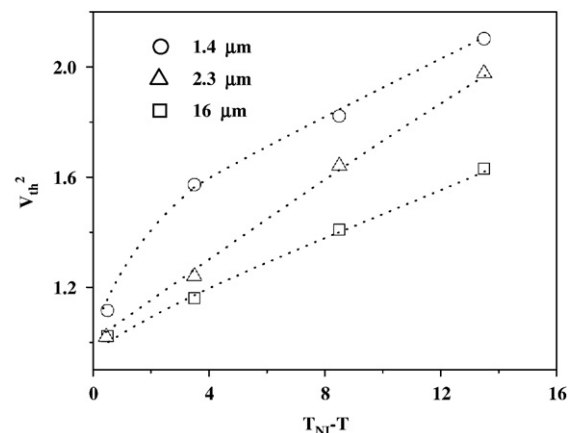

Fig. 8. V_{th}^2 at a few temperatures in three different cells of the 50 wt.%:50 wt.% mixtures of compound-2 and compound-3. Open circles (1.4 µm), open triangles (2.3 µm), open squares (16 µm). Dotted lines are drawn as guides to the eye. Frequency: 3037 Hz.

Table 1
Relative enhancement of orientational order parameter computed from birefringence and Freedericksz's threshold voltage.

Thicknesses of thin and thick cells	Compounds	T_{NI} in °C	$(T_{NI} - T)^0$	$\frac{\Delta n_{thin} - \Delta n_{thick}}{\Delta n_{thick}} \times 100$	$\frac{(V_{th}^2)_{thin} - (V_{th}^2)_{thick}}{(V_{th}^2)_{thick}} \times 100$
1.4 and 7 μm	Compound-1	55.5	12	6%	16%
1.5 and 14 μm	Compound-2	154.2	12	11%	43%
1.4 and 16 μm	Mixture of compound-2 and compound-3 (50:50 wt.%)	118.5	12	13%	30%

temperatures. More importantly the intensity varies gradually across the transition region in thin cell and the variation is comparatively steeper in the 16-μm cell. In order to compare the relative sharpness of the transition we estimate the slope by $\Delta I / \Delta V$ in the transition region. The magnitude of the slope of the transition in 1.4-μm cell is ~5 whereas it is ~30 in case of the 16-μm thick cell. We also measured the threshold voltage from the birefringence measurement in two different cell thicknesses namely 1.4 μm and 14 μm in compound-2. The birefringence was measured using a tilting compensator as a function of applied voltage as shown in Fig. 5. It is noticed that the birefringence in thinner cell (1.4 μm) is higher (6%) compared to the thick cell (14 μm). Similar enhancement of birefringence in the absence of a field was also reported by us on several other nematic liquid crystals and the origin of such enhancement is not completely understood [6].

In order to bring out the strong influence of cell thickness we show the variation of threshold voltage in two compounds and the mixture in Figs. 6–8. We plot V^2_{th} as a function of temperature instead of V_{th} and the reason will be discussed later. As we can see from Fig. 6, V^2_{th} is higher in 1.4-μm cell than that in 7-μm cell of the compound-1. V^2_{th}, measured in three different cells of the compound-2 are shown at a few temperatures in Fig. 7. Similarly V^2_{th} for the mixture is shown at a few temperatures in Fig. 8. It is observed that with decreasing cell thickness V^2_{th} is increased at a fixed temperature in all the systems. It is also noticed that due to the addition of banana molecules in the mixture, the threshold voltage measured in all the cells is increased significantly compared to that of the compound-2. The threshold voltage of the Freedericksz's transition is given by $V_{th} = \pi\sqrt{K_1 / \varepsilon_0 \Delta\varepsilon}$, where K_1 is the splay elastic constant and $\Delta\varepsilon$ is the dielectric anisotropy. In the mean field model the elastic constant, $K_1 \propto S^2$ where S is the orientational order parameter and the dielectric anisotropy $\Delta\varepsilon \propto S$. Therefore $V^2_{th} \propto S$. Thus V_{th}, can get enhanced in thinner cells due to the enhancement of orientational order parameter.

In order to estimate the enhancement in V_{th} in thin cells which can arise due to the enhancement of order parameter alone, we collect the Δn data from Ref. [6] at a fixed temperature below the transition of the above compounds and summarize in Table 1. In compound-1, V^2_{th} is ~16% larger in 1.4-μm cell than that in 7-μm cell at ~12° below T_{NI} whereas Δn is ~6% larger at the same temperature. In the compound-2, V^2_{th} is ~43% larger in 1.5 μm than that in 14-μm cell whereas Δn is ~11% larger at the same temperature. Similarly in the mixture V^2_{th} is 30% larger in 1.4-μm cell than that in 16-μm cell whereas Δn is ~13% larger at the same temperature. These discrepancies can be explained as follows. Though the liquid crystals are dielectric but they exhibit some pronounced ionic effect which are generally not observed in ordinary dielectric liquids. One of such effect is the space charge polarization which depends on concentration of ions, applied voltage and cell thickness. The source of ions in liquid crystal are not only residual impurities but probably also decomposition of the basic liquid crystal compounds. The transit time ($t = d^2 / \mu V$, where μ, is the mobility, V is the applied voltage and d is the cell thickness) of these ions in the thin cells is small and hence the ions can easily reach the appropriate electrodes when the field is reversed,

producing a space charge. This effect is more in the low frequency region. The space charges partially screen the external electric field. Therefore the effective field inside the thin cells is decreased. The large value of V^2_{th} in thin cell arises due to the screening effect of the space charge apart from the enhancement of S. It is also noticed from Table 1 that in the compound-1 and in the mixture the enhancement of V^2_{th} in thin cells is ~2.5 times larger than that of Δn whereas this is ~4 times larger in compound-2. In this compound the nematic phase occurs at high temperatures (105.5–154.2 °C). The density of ions increases with the increase of temperatures and as a result the screening effect is increased. The larger threshold voltage in case of the mixture with respect to the pure compound may indicate the stiffening of splay elastic constant in the mixtures due to the addition of the banana-shaped molecules. The transition is ~6 times sharper in thick cell (16 μm) compared to the thinner (1.4 μm) one. This effect may arise due to the stronger surface anchoring energy in thinner cell compared to the thicker one.

Finally we want to comment on the effect of addition of bent core molecules in the mixtures. $V^2_{th}(\sim S)$ in the mixture is larger compared to the pure compound in all the cells indicating that the orientational order parameter is increased. However, the understanding of mutual orientations of two differently shaped molecules is important to explain the results further. At present we are pursuing some investigations to understand the mutual orientations.

4. Conclusion

Our experiments show that the Freedericksz's transition in thin (~1–3 μm) cell is much less steeper compared to the thicker cells (~7–16 μm). The threshold voltage of nematic liquid crystals in thin cells is enhanced considerably with respect to that in thick cells for materials with positive dielectric anisotropy. Part of the enhancement arises due to the enhancement of orientational order parameter in thin cells. The space charge polarization exhibits strong screening effect in thin cells at higher temperatures as a result the nonuniform electric field arising from the space charge effects enhances the Freedericksz's threshold voltage. Liquid crystals which are used in very thin display devices have to be highly purified to minimize the effect of space charge polarization to the electro-optic characteristics.

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