

Liquid Crystals

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Alignment of unconventional nematic liquid crystals

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Planar (homogeneous) and homeotropic alignment of some unconventional nematic liquid crystals with both positive ($\Delta \epsilon > 0$) and negative ($\Delta \epsilon < 0$) dielectric anisotropies are studied on planar (AL-1254) and homeotropic (JALS-204, SE-1211) alignment layers. Irrespective of the sign of dielectric anisotropy and molecular structure, uniform planar alignment of the director is observed in Al-1254 coated cells whereas no stable homeotropic alignment of bent-core compounds with $\Delta \epsilon < 0$ is observed in cells coated with JALS-204 or SE-1211. Alignment properties of these two types ($\Delta \epsilon > 0$ and $\Delta \epsilon < 0$) of unconventional nematic liquid crystals are explained considering the steric, short-range dipolar and long-range van der Waals interactions at the interface. A bent-core compound ($\Delta \epsilon < 0$) which has no higher-order liquid crystalline phase except for the nematic exhibits a narrow temperature range of homeotropic state (on JALS-204) while cooling from the isotropic phase followed by an anchoring transition to a planar state as the temperature is further lowered. The measured polar anchoring energy of this compound on JALS-204 is significantly lower than many known calamitic liquid crystals and is responsible to the anchoring transition observed.

Keywords: bent-core liquid crystals; planar alignment; homeotropic alignment; dielectric anisotropy

1. Introduction

Uniform alignment of the liquid crystal director (denoted by \hat{n} , the average orientation of the long axes of the molecules) is technologically very important. It is also essential for any quantitative measurements of various physical properties. Commonly two categories of alignment layer are used, which promotes either planar or homeotropic alignment of the director. These alignment layers are very stable in the sense that the director orientation does not change with time and temperature. Most of the liquid crystals with rod-like molecules (calamitic) can be aligned easily in the nematic phase both in planar and homeotropic cells, and the majority of the liquid crystal display devices (LCDs) make use of these alignment layers.

The unconventional nematic liquid crystals so far have not been used in LCDs. Nevertheless, recently bent-core nematics has drawn much attention because of their many uncommon physical properties such as an emerging biaxial nematic phase [1], unconventional electroconvection [2, 3], flexoelectric [4] and Kerr effects [5], sign inversion of elastic anisotropy [6], larger shear and rotational viscosities [7], low pretilt angle [8], when compared to calamitic liquid crystals. In many of these measurements the director is aligned in the planar cell uniformly. There are some reports that bent-core nematic liquid crystals do not align homeotropically with the conventional alignment layers [9, 10]. However, the effect of dielectric anisotropy, and in particular the role of the dipole moment and steric effect, on the alignment behaviour has not been investigated. In this paper we report that most of the unconventional nematic liquid crystals, irrespective of their sign of dielectric anisotropy and molecular structure, can be aligned in planar cells and bentcore nematic liquid crystals with negative dielectric anisotropy in general cannot be aligned homeotropically. Further, we show that a bent-core nematic which has no higher-order liquid crystalline phase at lower temperature exhibits a continuous anchoring transition from homeotropic to planar on JALS-204 as the temperature is reduced. We show that the polar anchoring energy for bent-core liquid crystals with negative dielectric anisotropy is low and could be responsible for the inability to provide stable homeotropic alignment.

2. Experimental details

We used AL-1254 as planar and JALS-204 and SE-1211 as homeotropic alignment layers. Indium tin oxide (ITO) coated glass plates were spin coated with AL-1254 and cured at 180°C for one hour

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for planar alignment. Similarly JALS-204 and SE-1211 spin coated ITO glass plates were cured at 200°C and 180°C respectively for one hour for the homeotropic alignment. The AL-1254 coated plates were rubbed in an antiparallel direction to obtain planar alignment of the liquid crystal director. An ultraviolet (UV) curable adhesive mixed with $\sim 5 \,\mu$ m spacer was used to make cells. The sample was filled in the isotropic phase and cooled slowly across the nematic–isotropic transition. Texture observation was made by using a polarising optical microscope (Olympus BX-51) connected with a Instec hotstage (Instec Inc.) and digital camera.

The polar anchoring energy of the compound with $\Delta \epsilon = (\epsilon_{\parallel} - \epsilon_{\perp}) < 0$ was measured in the homeotropic cell by using a technique proposed by Nie et al. [11]. When the applied voltage exceeds a threshold, the liquid crystal director in the bulk is reoriented and the reorientation at the surface occurs at much higher voltage due to the finite surface anchoring. Nie et al. [11] showed that the anchoring energy in a homeotropic cell can be measured from the voltage-dependent retardation data when other material parameters are known. The sample capacitance and the retardation were measured simultaneously as a function of voltage. The sample was placed between two crossed Glan-Thompson polarisers and a He-Ne laser (632.8 nm) was used as a light source. A photoelastic modulator (PEM-100, Hinds Instruments) was placed between the polariser and the sample making the retardation axis parallel to the rubbing direction. The first and the second harmonics of the optical intensity were measured by using a photodiode and lock-in amplifier (SRS-830). The retardation was estimated from the measured intensities. A LCR meter (Agilent E4980A) at 4111 Hz was used to measure the capacitance. All the measurements were controlled with the help of a computer. The anchoring strength (W) was estimated from the following equation [11]:

$$\frac{R}{R_0} = -\frac{1}{CV} \frac{\xi}{\Delta n} I(b, \gamma, \nu, \pi/2) + (1 + \frac{2K_{33}}{Wd}), \quad (1)$$

where

$$\xi = (\epsilon_0 \epsilon_\perp A/d) \pi \sqrt{K_{33}/\Delta \epsilon}$$

and

$$I(b, \gamma, \nu, \theta) = \frac{2}{\pi} \int_0^\theta \frac{1 - \nu + (1 - \nu)^{1/2}}{1 - \nu \cos^2 \phi + (1 - \nu \cos^2 \phi)^{1/2}}$$
$$\frac{(1 + \gamma \cos^2 \phi)^2 (1 + b \sin^2 \phi)^{1/2}}{\sin \phi} d\phi.$$

 R_0 is the maximum retardation and R/R_0 is the normalised phase retardation. C, V, d and A are the sample capacitance, applied voltage, cell thickness and electrode area. In Equation (1), $v = (n_e^2 - n_o^2)/n_e^2$, where n_e and n_o are the extraordinary and ordinary refractive indices, $\gamma = (K_{11} - K_{33})/K_{33}$ and b = $(\Delta\epsilon)/\epsilon_0\epsilon_{\perp}$. K_{33} is the bend elastic constant and is measured from the Freedericksz threshold voltage (V_{th}) . $(\xi/\Delta n)I(b, \gamma, v, \pi/2)$ depends on the material parameters and is independent of voltage. The polar anchoring energy, W, is determined from the intercept of the plot R/R_0 as a function of $1/CV_{rms}$.

3. Results and discission

Several compounds with unconventional molecular structures and in various temperature ranges of the nematic phase were chosen for the experiment. The textures obtained in the planar and homeotropic cells for liquid crystals with positive dielectric anisotropy ($\Delta \epsilon = \epsilon_{||} - \epsilon_{\perp} > 0$) are shown in Figures 1 and 2. The dielectric anisotropy of the compound in Figure 1(a) is $\Delta \epsilon \sim 1$ at T_{NI} – $T = 20^{\circ}$ and exhibits a large temperature range of the nematic phase. Various physical measurements on this compound have already been reported [6, 12–15]. Uniform planar and homeotropic alignments of the nematic director are obtained in planar (Figure 1(b)) and homeotropic (Figure 1(c)) cells, respectively. The homeotropic state is very stable in the entire nematic phase except tilting of the director at much lower temperature in some parts of the sample is observed as reported earlier on various other homeotropic alignment layers [12, 15]. The molecular structure of the compound in Figure 1(d) is hockey-stick shaped and exhibits a positive dielectric anisotropy ($\Delta \epsilon \sim 1.6$ at $T_{NI} - T = 4^{\circ}$). The detailed physical measurement on this compound will be reported elsewhere [16]. Uniform planar and dark homeotropic states are observed in the planar (Figure 1(e)) and homeotropic (Figure 1(f)) cells, respectively. The compound in Figure 2(a) is composed of branched molecules and exhibits positive dielectric anisotropy ($\Delta \epsilon = 0.9$ at $T_{NI} - T = 4^{\circ}$) [17]. The molecule appears to be T-shaped although the optimised structure of a single molecule shows that the different parts of the molecule are not in the same plane [17]. It also shows good planar (Figure 2(b)) and homeotropic (Figure 2(c)) alignment on the respective alignment layers.

The textures of compounds with $\Delta \epsilon < 0$ are shown in Figures 3 and 4. The dielectric anisotropy of the compound in Figure 3(a) is $\Delta \epsilon \sim -1$ at $T_{NI} - T = 3^{\circ}$ [8]. Uniform planar alignment of the director is observed in planar cells (Figure 3(b)) in this



Figure 1. (a) Molecular structure and phase transition temperatures of the compound ($\Delta \epsilon > 0$). Textures observed in the (b) planar and (c) homeotropic cells at a temperature of 170°C. Cell thickness: 6.9 µm and 8 µm for the planar and homeotropic cells, respectively. (d) Molecular structure and phase transition temperatures of the compound ($\Delta \epsilon > 0$) with hockey-stick shaped molecules. Textures observed in (e) planar and (f) homeotropic cells at a temperature of 120°C. Arrows in (b) and (e) denote the rubbing directions. Cell thickness: 7.1 µm and 4.2 µm for the planar and homeotropic cells, respectively (colour version online).

JALS-204

compound. In homeotropic cells (Figure 3(c) and 3(d)) the textures appear mostly dark and exhibit no schlieren texture suggesting that the sample is partially homeotropic. The molecule in Figure 3(e) has a larger number of phenyl rings and exhibits a high-temperature nematic phase. No Freedericksz

AL-1254

transition in the nematic phase was observed in a planar cell suggesting that the dielectric anisotropy is negative. The director is uniformly aligned in a planar cell (Figure 3(f)) whereas no homeotropic alignment is observed in homeotropic cells (Figure 3(g) and 3(h)). In Figure 4 we show the textures obtained



Figure 2. (a) Molecular structure and phase transition temperatures of the compound ($\Delta \epsilon > 0$) with branched (T-shaped) molecules. Textures observed in the (b) planar and (c) homeotropic cells at a temperature of 117°C. Cell thickness: 5.2 µm and 6 µm for the planar and homeotropic cells, respectively. The arrow in (b) denotes the rubbing direction (colour version online).

in a relatively low temperature and well characterised bent-core nematic liquid crystal which has no higher order liquid crystalline phase at lower temperature [2, 3, 18–20]. It shows uniform planar alignment in a planar cell (Figure 4(b)) as seen in other samples. No homeotropic alignment is observed in cells coated with SE-1211 (Figure 4(c)). In JALS-204 coated cells, initially they show good homeotropic alignment up to 70° C (Figures 4d(i)) when the sample is cooled from the isotropic phase and exhibit a continuous anchoring transition (Figures 4(e)(ii)-4(e)(iv)) as the sample is further cooled down, finally becoming almost planar at 51°C. A similar continuous anchoring transition of a calamitic nematic mixture (E7) on perfluoropolymer was reported by Patel et al. [21]. Thus from the texture observation of the compounds studied it appears that unconventional nematic liquid crystals show planar alignment irrespective of the sign of the dielectric anisotropy. The compound with positive dielectric anisotropy can be aligned uniformly in homeotropic cells whereas bent-core nematic materials with negative dielectric anisotropy in general cannot be aligned in homeotropic cells.

There are several interfacial forces that are responsible for the orientation of the liquid crystal molecules on the substrate surfaces, such as steric interactions and short-range dipolar and long-range van der Waals interactions. Steric interactions prefer planar alignment of the director [22, 23]. Longrange van der Waals interactions prefer the large polarisability direction (here the bow axis denoted by P in Figure 5) parallel to the substrate surface [24, 25]. Thus both the steric and van der Waals interactions in bent-core liquid crystals always prefer planar alignment of the director. The short-range dipolar interactions tend to make dipoles normal to the interface to maximise their interaction with the charges at the surface and have an opposite



Figure 3. (a) Molecular structure and phase transition temperatures of the compound ($\Delta \epsilon < 0$). Textures observed in the (b) planar and (c),(d) homeotropic cells at a temperature of 117°C. Cell thickness: 5.4 µm for planar and 5.7, 5.3 µm for homeotropic cells, respectively. (e) Molecular structure and phase transition temperature of the compound with n = 7 ($\Delta \epsilon < 0$). M denotes an unidentified smectic phase. Textures observed in the (f) planar and (g), (h) homeotropic cells at a temperature of 207°C. Cell thickness: 5.3 µm for the planar and 4.5, 5 µm for the homeotropic cells, respectively. Arrows in (b) and (f) denote the rubbing directions.

alignment effect depending on the strength and orientation of the resultant dipole moment with respect to the long molecular axis. They tend to align the long molecular axis (P) perpendicular to the surface where the direction of the resultant dipole moment (denoted by D in Figure 5(a)) is parallel to P. On the other hand, they tend to orient the bow axis (P) parallel to the substrate where the direction of dipole moment (D) is perpendicular to P. In the case of compounds with $\Delta \epsilon > 0$, D is parallel to P and has opposite alignment effect. However, if the dipolar interactions are relatively strong compared to that of the long-range van der Waals interactions they can lead to homeotropic alignment of the bent-core molecules as schematically shown in Figure 5(c). In the case of bent-core compounds with $\Delta \epsilon < 0$, D is parallel to the arrow axis, i.e. almost perpendicular to P (Figure 5(b)) and both D and P have similar alignment effects, i.e. planar alignment of the molecules (Figure 5(d)). As a result, dielectrically-negative-type bent-core nematic compounds in general do not exhibit homeotropic alignment. The anchoring transition in Figure 4 can be considered as a result of competition between these two major interactions as



Figure 4. (a) Molecular structure and phase transition temperatures of the compound ($\Delta \epsilon < 0$). Textures observed in the (b) planar (AL-1254) and (c) homeotropic (SE-1211) cells at 72°C. Textures observed in the homeotropic cell (JALS-204) at various temperatures when the rubbing direction is parallel ((d)(i)–(d)(iv)) and rotated by 45° ((e)(i)–(e)(iv)) with respect to the polariser. Arrows in the figure denote the rubbing directions. Cell thickness: 7.1 µm for the planar and 4.2, 5.2 µm for the homeotropic cells, respectively (colour version online).

discussed. Since most of the bent-core liquid crystals possess several phenyl rings, the polarisability is large and is strongly temperature dependent. Because of increased polarisability at lower temperatures the van der Waals interactions can dominate over the dipolar interactions and an anchoring transition can occur.

The compound in Figure 4(a) shows an unstable homeotropic state (from 75.3°C to 70°C) in the sense that below 70°C it exhibits a continuous anchoring transition to the planar state. We measured the anchoring energy exploiting the homeotropic alignment of this compound. Retardation (R) and capacitance (C) were measured simultaneously to measure the anchoring energy (W) at a particular temperature. The variation of retardation and capacitance as a function of applied voltage is shown in Figure 6(a) and Figure 6(b), respectively. Below the Freedricksz threshold voltage ($V_{th} = 0.47V$) the sample retardation is zero (homeotropic state) and the director tilts as the voltage is increased and finally becomes almost planar at higher voltages. The bend elastic constant was calculated by using the relation $K_{33} = (V_{th}/\pi)^2 \Delta \epsilon \epsilon_o$ where $\Delta \epsilon$ is the dielectric anisotropy. At 72°C, $\epsilon_{\parallel} = 5.5$ and



Figure 5. Schematic representation of bent-core molecules. The direction of the resultant dipole moment and the large polarisability directions are denoted by D and P, respectively. (a) Both D and P are parallel in materials with $\Delta \epsilon > 0$. (b) D and P are mutually perpendicular in materials with $\Delta \epsilon < 0$. (c) Homeotropic alignment of the director with $\Delta \epsilon > 0$. (d) Planar alignment of the director with $\Delta \epsilon < 0$.



Figure 6. (a) Retardation as a function of applied voltage measured in the homeotropic cell (at 72°C). (b) Sample capacitance as a function of applied voltage at 72°C. (c) Normalised retardation R/R_o as a function of $1/CV_{rms}$ at 72°C. The continuous line is the theoretical fit by Equation (1). Cell thickness: 6.1 μ m.

 $\epsilon_{\perp} = 6.9$, which were measured from the extrapolation of C with $1/V_{rms}$. The estimated K_{33} from the present Freedericksz threshold measurement is 0.3×10^{-12} N, which is comparable in magnitude with the reported values [26]. A similar value of K_{33} near the nematic-isotropic transition has also been reported for other bent-core nematic liquid crystals [6]. The variation of the normalised retardation (R/R_o) as a function of $1/CV_{rms}$ is shown in Figure 6(c). The experimental data were fitted to the theoretical Equation (1) by using a least-squares fitting method to estimate the intercept. The anchoring energy, W, was estimated to be $1.4 \times 10^{-5} \text{ J/m}^2$, which is significantly low compared to many known calamitic liquid crystals. The anchoring transition observed in this compound could be attributed to the low anchoring energy.

4. Conclusion

We have studied alignment properties of some bent-core, hockey-stick and T-shaped nematic liquid crystals with both positive and negative dielectric anisotropy. We found that these unconventional nematics with both positive and negative dielectric anisotropy can be aligned easily in planar cells whereas bent-core materials with negative dielectric anisotropy in general cannot be aligned homeotropically. The experimental results are explained by considering the steric, short-range dipolar and van der Waals interactions. The polar anchoring energy of a material with negative dielectric anisotropy is very small and perhaps leads to the anchoring transition as observed. Further, it will be interesting to study and measure the surface-related properties of bentcore nematic compounds with different shapes on various other alignment layers.

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