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Liquid Crystals

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Liquid crystal alignment on isotropic submicro-textured surfaces of homeotropic-planar states

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The equilibrium configuration of liquid crystal molecules in contact with a periodically patterned substrate has been theoretically analysed considering the variation of the order parameter at the interface, and a first order transition was predicted for a critical periodicity, or induced by temperature or an electric field. In this work, we experimentally investigated the equilibrium configuration of the director for a submicron patterned substrate, alternating homeotropic and random planar regions in the substrate. Experimentally, it was possible to access a starting point, theoretically close to the first order transition, however no transition was observed either on varying the temperature or on applying an electric field. The possible reasons for the non-observation of the phase transition are discussed.

1. Introduction

The problem of effective liquid crystal (LC) alignment induced by non-homogeneous substrates or cells with hybrid alignment, was theoretically analysed taking into account the director spatial variation, but neglecting the variation of the order parameter at the interface [1, 2]. It is equivalent to consider an infinite correlation length in the continuum Frank–Oseen elastic theory, resulting in the homogenization of the director orientation [3]. A more realistic analysis was done by Qian and Sheng [4, 5], considering the order parameter variation at the interface of the nematic medium and the substrate. Using the Landau–de Gennes formalism, it was suggested that the order parameter variation, near the surface, results in an elastic relaxation effect, which according to numerical simulations, leads to new orientational states and bulk orientational transitions. This would be possible for a patterned substrate, with periodic homeotropic and planar alignment conditions. The orientational transitions could be induced by changes in the periodicity, temperature or by applying a voltage to the LC cell.

In a recent paper, the orientation of a nematic liquid crystal in contact with a binary isotropic substrate was described by Lee and Clark [6]. The surface area was divided into two distinct regions of homeotropic (HO) and random planar (RP) alignment induced to the LC. The homeotropic alignment was obtained by coating a glass plate with octadecyltriethoxysilane

(OTE) self-assembled monolayers (SAMs). Then, some regions of the OTE film are exposed to unpolarized UV light, which affects the bond between the silane and the alkyl tail. In these regions the film can be removed, resulting in a random planar alignment of the LC molecules. The step in the surface relief at the boundary of the irradiated and non-irradiated OTE film was determined by atomic force microscopy and is less than 2.5 nm high (as expected for complete SAM removal), being small enough to eliminate the possibility of LC alignment due to surface topography [7].

In this work, we experimentally investigate the orientational states induced by a patterned substrate, combining homeotropic and random planar periodic stripes. The possibility of bulk phase transitions was also investigated by varying some parameters, such as the surface periodicity and the LC temperature. To obtain a patterned surface, an interference pattern was generated on the substrate coated with OTE self-assembled monolayers. The influence of the electric field on the orientational states was also investigated by applying a voltage to the nematic cell.

2. Theory

In this section we will present the main assumptions and results derived from the analysis of the different orientational states induced by a patterned substrate [4, 5]. The patterned substrate consists of homeotropic and random planar regions, with a periodicity P , along the y -axis, with the substrate located at $z=0$; the tilt angle θ , is measured from the substrate. The free energy of the

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nematic liquid crystal is written as a sum of the bulk free energy and surface free energy. In the formalism of Landau–de Gennes, the bulk free energy is written as an expansion in a power series of the tensor order parameter Q . The surface free energy is expressed in terms of a surface coupling constant $G(x)$, which is equal to $-G_n$ in the homeotropic region and G_p in the planar region, where $G_p > 0$. The total free energy, F , can be expressed as a dimensionless parameter $\Psi = F/(\Delta^3 B^4/C^3)$, where B and C are the phenomenological constants used in the Landau–de Gennes expansion of the free energy, and Δ is the intrinsic elastic correlation length of the nematic phase; the dimensionless surface coupling is given by $g = G/(\Delta B^3/C^2)$.

Numerical minimization of the free energy expression is carried out to determine the equilibrium configurations of the liquid crystal. The equilibrium bulk orientation arises from the competition between the elastic energy in the boundary layer and the surface alignment potential, depending on the relation between the periodicity P and Δ . For $P \geq 10\Delta$, the configuration of the director can be divided into two regions; (i) the boundary close to the surface, with typical thickness $\approx 2P$, characterized by strong spatial variation of the order parameter along the x -axis, and (ii) the bulk, extending from $z \geq 2P$ to ∞ .

A homogeneous state, denoted YZ, is stable when the periodicity is large so that the elastic energy in the boundary layer is small compared with the surface alignment potential. The tilt angle is approximately 40° , in the limit of large P , and decreases continuously with decreasing P , up to a critical value periodicity P_c , with $\theta = 7^\circ$. Then a first order transition takes place, leading to a homogeneous state, denoted X state, with the director parallel to the x -axis. The configuration of the director in the different bulk states is schematically represented in figure 1, reproduced from [4, 5]. The critical periodicity is $P_c = 47.3\Delta$, for a fixed temperature

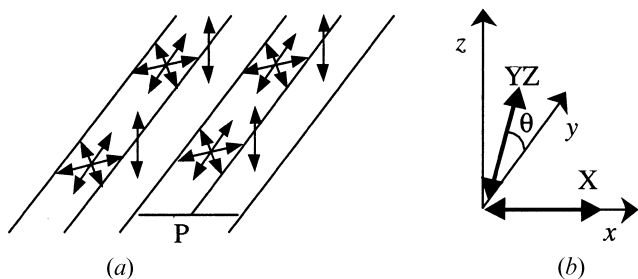


Figure 1. (a) Schematic illustration of the texture stripe pattern with alternating HO and RP alignment conditions. The directors of the two bulk orientational states are shown at right. (b) The director configurations of the YZ state and the X state in the transition boundary layer. This figure comes from [4].

unit, $t = -1.0$, where $t = aC(T - T_c)/B^2$, a is a positive constant and T_c is the nematic–isotropic phase transition temperature. For the 5CB liquid crystal, $\Delta = 40 \text{ \AA} \Rightarrow P_c = 189.2 \text{ nm}$, for a fixed temperature $T = 31.09^\circ\text{C}$. Keeping fixed the periodicity $P \approx 50\Delta$, the same transition is obtained as a function of the temperature. Considering $P = 190 \text{ nm}$, the transition takes place for $t = -1.017$, which corresponds to $T = 31.01^\circ\text{C}$. From the relation between P and T (for P around 50Δ) one observes that if P is increased, the first order phase transition can be obtained due to the reduction of T , and *vice versa*. This means that by varying these two parameters one can find the bulk phase transition, from the YZ to the X state, however no calculations were carried out for temperatures far from the transition temperature, since the Landau–de Gennes free energy expansion cannot be applied.

The same transition can be induced by an electric field applied perpendicular to the substrate, which is in favour of the X state. When the system is in the YZ state, the field-induced free energy can be comparable to the difference between the energy stored in the boundary layer of the X and YZ states, and this occurs for a critical voltage, V_c . If V_c is lower than the critical Fréedericksz transition voltage, V_F , one can assume that the configuration of the director in the YZ state is not affected by the electric field; a first order approximation can then be used to determine V_c , which is found to be proportional to $d^{\frac{1}{2}}$. Therefore, there exists a maximum cell thickness, d_F , for which this approximation is valid, with $V_c < V_F$. The critical thickness is large enough to be experimentally accessible, starting with the system in the YZ state, but close to the first order transition. For $P = 50.0\Delta$ and $t = -1.0$ the maximum thickness is found to be $d_F = 2.2 \times 10^3 \Delta$. For the liquid crystal 5CB, ($V_F = 3.0 \text{ V}$); this situation corresponds to a periodicity $P = 200 \text{ nm}$, $d_c = 6.6 \mu\text{m}$ with a critical voltage $V_c = 2.4 \text{ V}$. Nevertheless, for strong anchoring at the boundary layer, the field coherence length is smaller than the thickness of the boundary layer and the field-induced transition is not possible. These predictions resulted from simulation where the coupling constant parameter was assumed to be $G = 6.14 \times 10^{-4} \text{ J m}^{-2}$, and the corresponding dimensionless surface coupling parameters were assumed to be $g_n = 0.05$ and $g_p = 0.08$.

3. Experimental

The OTE [$\text{CH}_3(\text{CH}_2)_{17}\text{Si}(\text{OC}_2\text{H}_5)_3$] was acquired from United Chemical Technologies, Inc., Bristol, PA. The OTE self-assembled monolayer was prepared from a solution of 5% (vol) of OTE in toluene with 0.5% (vol) of decylamine as catalyst [8]. We used glass microscope

slides as substrates, which were carefully cleaned and dried before the film deposition. The glass plates were sonicated in the solution for 60 min, followed by a 30 min soak in the same solution. The plates were then dried at 80°C for 15 min for total solvent evaporation. This process leads to a monolayer of OTE attached with the hydrophilic heads at the glass surface. The same treatment was given to glass plates coated with indium tin oxide (ITO).

The absorption spectrum of the pure OTE was measured with a diode array spectrophotometer (Hewlett Packard/HP, 8452A) and a strong absorption peak was observed around 226 nm. The UV irradiation of the film was performed by a 1000 W Hg(Xe) arc lamp (Oriental Corporation, 6293), which presents a wide emission spectrum with a well pronounced peak around 245 nm. This peak corresponds to 3.0% of the total lamp irradiance and is responsible for the effective OTE irradiation. The sample was positioned at a distance of 20 cm from the collimating lens with effective incident power of 200 mW cm⁻² (coming from the 245 nm peak) for 20 min exposure time. Immediately after the irradiation the sample was rinsed with ethanol to remove residues.

Textured patterns were obtained by using an interferometric setup, as shown in figure 2, where the periodicity (P) could be controlled by $P = \lambda / (2 \sin \theta)$. Considering $\lambda \approx 245$ nm, we obtained $P \approx 213$ nm for $\theta = 35^\circ$ (35° is the limiting angle in the experiment). It is important to note that the lamp is not monochromatic, and in fact other wavelengths may contribute to the interference pattern on the substrate.

In the experiments, the LC 5CB (Merck) was used without further purification and it was inserted into the cells constructed with the treated substrates at room temperature, i.e. with the sample in the nematic phase. The LC texture was captured with a digital camera coupled to a polarized microscope.

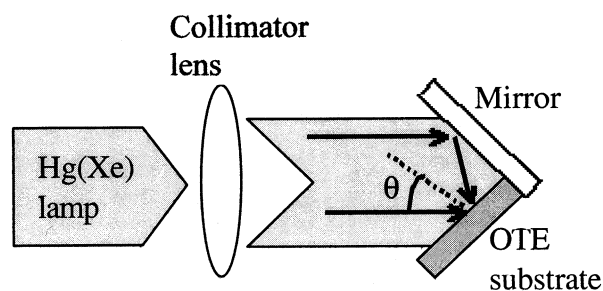


Figure 2. Experimental set-up used for OTE irradiation, generating interference patterns.

4. Results and discussion

Initially, we investigated the irradiation efficiency of the UV lamp by irradiating an OTE film using a metal mask to allow illumination in specific regions, separated by 100 μ m. Then periodic patterns of irradiated and non-irradiated stripes were recorded on the substrate. A cell was mounted to have the direction of the stripes in the second boundary layer perpendicular to the first. The resulting texture for a 23 μ m thick cell is shown in figure 3, between crossed polarizers. The dark squares correspond to the superposition of non-irradiated areas, or a homeotropic alignment; the bright areas correspond to irradiated areas, or random planar orientation.

The identification of the YZ state was made by preparing cells (23 μ m) in which one of the boundary surfaces was the patterned substrate and the second was coated with lecithine to induce homeotropic alignment. Two different periodicities were recorded in the patterned substrate, 358 and 705 nm, and the texture was observed in a polarizing microscope (figure 4). By rotating the sample it was seen that the orientation is homogeneous with the director oriented parallel to the y-axis (parallel to the direction of the stripes), however the light is not completely extinguished, indicating that there is a tilt angle.

The anchoring strength of the patterned substrate was estimated using the hybrid twisted cell method [9]. The cell was built using spacers of 23 μ m and in the second boundary surface (surface B) a PVA rubbed film, with the rubbing direction making an angle of 90° with respect to the direction of the stripes in the first boundary surface (surface A). The PVA rubbed film is

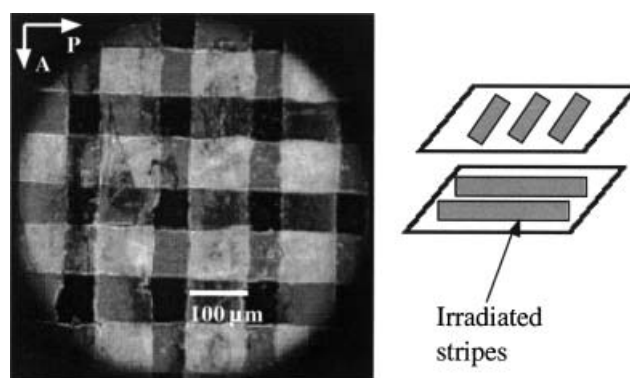


Figure 3. LC texture between crossed polarizers. The cell was constructed with crossed plates of periodically irradiated stripes of 100 μ m width, as shown in the inset. The bright and black squares indicate superimposing irradiated and non-irradiated regions, respectively.

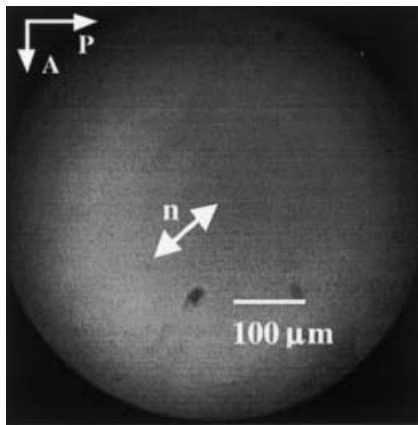


Figure 4. YZ orientational state induced by homeo-planar patterned substrate with $P \approx 358$ nm.

well known to induce a strong azimuthal anchoring ($W\phi \approx 1.5 \times 10^{-5} \text{ J m}^{-2}$). For comparison purposes, surface A was divided into two regions, one recorded with the interference pattern and the other protected from UV light. With $P \approx 213$ nm, a total twist of the director was observed at the interferometric patterned area and a planar alignment (induced by the rubbed PVA) on the non-irradiated part. In figures 5(a) and 5(b), one can see that the twist region (irradiated part) remains bright on rotating the sample 45° between crossed polarizers, whereas, in the non-irradiated part the LC orientation is dominated by the PVA, being dark when \mathbf{n} of the PVA is parallel to one of the polarizers and bright when it is at 45° . These results indicate that the anchoring energy at the irradiated area is strong and of the same order of the rubbed PVA film. The same result was obtained for periodicities $P \approx 358$ and 705 nm. For the periodicities used in these experiments, it was observed that the bulk orientation corresponds to YZ state.

In the sequence, the possibility of a phase transition from the YZ to the X state was investigated by decreasing the periodicity to $P \approx 213$ nm, which is the limiting value for the experimental set-up described above. Since the critical periodicity is below this value, it was not possible to investigate the phase transition by further decreasing the periodicity. However, with this starting condition, $P \approx 50\Delta$, the transition can be induced by temperature variation and, as discussed above for the 5CB liquid crystal, it would occur for $T \approx 31^\circ\text{C}$. For this purpose, the same hybrid cell as presented in figure 5 was used. The phase transition from the YZ to the X state should be accompanied by a transition from a homogeneous alignment in the patterned substrate area to a planar orientation. The temperature was changed over approximately the whole of the nematic range of this LC sample (24 to 34°C) and

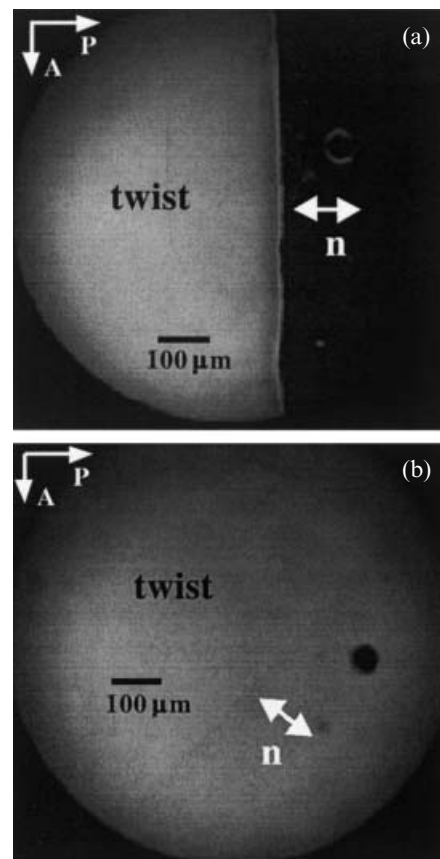


Figure 5. LC texture in a hybrid cell between crossed polarizers, showing the interface of irradiated and non-irradiated area. One of the cell plates was irradiated with the interferometric set-up ($P = 213$ nm), and the other is a unidirectional PVA rubbed surface: (a) parallel to the analyser, and (b) 45° from the analyser and polarizer.

no orientational transition was observed. For this experiment, the LC cell was placed on a hot stage, allowing temperature control with a 0.05°C accuracy.

The possibility of a phase transition induced by the electric field was also investigated. For this purpose a $6\mu\text{m}$ thick cell was prepared, and the OTE film was drawn on ITO-coated glass plates. A twisted cell was mounted with a periodic pattern with $P \approx 213$ nm at one side and a PVA rubbed film on the other. Keeping the temperature constant at 31.1°C , the voltage was applied between the two substrates, with a continuous current source. The voltage was increased from $V = 1.0$ V up to the Fréederickzs transition threshold ($V = 3.0$ V), with a precision of ± 0.1 V; however no transition to the X state was observed.

According to the predictions discussed in §2, we expected to observe the transition from the YZ to the X state either on varying the temperature or by applying an electric field, since the starting point was not far

from the transition conditions. The fact that no transition was observed is probably related to the surface coupling on the patterned substrate. In the analysis presented in [4, 5], the coupling constant G was assumed to be $6.14 \times 10^{-4} \text{ J m}^{-2}$, and in our experiments we estimated the anchoring strength on the patterned substrate to be $1.5 \times 10^{-5} \text{ J m}^{-2}$; but this corresponds to an estimation of a minimum value, it could be larger than this. The analysis carried out in [4, 5], considers that in the YZ state the elastic energy stored in the boundary layer is similar to the surface energy in the X state, and the electric field can supply the difference of energy to induce the phase transition. This situation corresponds to an effective weak zenithal anchoring. Therefore, in our experiments, it is possible that the effective anchoring strength on the patterned substrate does not fulfill this condition.

5. Conclusions

It was shown that using the OTE self-assembled monolayer and the set-up described in section 3 it was possible to obtain patterned surfaces with periodicities in the micron and submicron scale. The use of these patterned substrates allowed the identification of the YZ state for different periodicities, and we investigated the possibility of a phase transition to the X state, on varying the temperature and on applying an electric field. Experimentally, it was possible to access a starting point, theoretically close to the first order transition, however no transition was observed either on varying the temperature or on applying the electric field. It is

possible that in these experiments, the value of the coupling constants, g_n and g_p , are very different from that used in the simulations made by Quian and Sheng [4, 5]. An effective larger anchoring strength would favour the YZ state and then, the difficult experimental access to the X state. A complete view of this transition would be achieved considering the effects of g_n and g_p on the equilibrium configurations. In addition, a quantitative description of the tilt angle (θ) variation in our experiments could help in estimating the distance from the transition point, however unfortunately it was not possible in this laboratory.

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