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Observation of bubble domains at the cholesteric to homeotropic-nematic transition in a confined chiral nematic liquid crystal

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We report observations on a chiral nematic liquid crystal confined in a plane cell treated for homeotropic alignment. The characteristics of the specimen investigated are such that at room temperature a uniform homeotropic alignment is observed and at higher temperatures the material displays the helical structure. It is observed that the changeover between homeotropic and helical textures does not occur at one single threshold. There are two thresholds, between which there is an intermediate texture where small circular domains are observed. These domains may be regarded as circular analogues of the plane solitons predicted by previous authors.

1. Introduction

The frustration of a chiral nematic liquid crystal subjected to an electric or a magnetic field is a classic problem that can be solved exactly [1, 2]. It is found that there is a threshold value of the field at which the texture of the material switches between a uniform alignment parallel to the applied field and the helical structure. Using a more sophisticated analysis, Kamien and Selinger [3] have shown that between the two extreme frustrated textures there is a regime where inversion domain walls or solitons can be formed in the material.

A cholesteric liquid crystal material can also be frustrated when it is confined in a container with at least one dimension of the order of or smaller than the bulk pitch of the material [4, 5]. In the case of a chiral nematic confined in a plane cell treated for homeotropic alignment one could make an analogy with the chiral nematic in a field. An effective field $\mathbf{H}_{\text{eff}}$ may be defined to describe the effect that surface anchoring has at a given point in the bulk. A formal definition of such an effective field and the corresponding susceptibility is not straightforward. This effective field will depend on the elastic constants of the material, which are themselves dependent on the temperature. The field will not be uniform across the specimen: it will be minimum in the middle of the cell. Nevertheless, one may say that as this effective field is decreased in a homeotropically aligned specimen there will be a threshold value at which the helical structure is restored, first in the middle of the cell and then to the entire specimen. The starting point for the analysis of the texture adopted by the material is to write the Frank free energy density of spatial distortion of the director in the applied field [3, 6]:

$$f=\frac{1}{2}k_1(\nabla.\mathbf{n})^2+\frac{1}{2}k_2(\mathbf{n}.\nabla\times\mathbf{n}+q_o)^2+\frac{1}{2}k_3(\mathbf{n}\times\nabla\times\mathbf{n})^2-\chi[\mathbf{H}_{\text{eff}}.\mathbf{n}]^2 \quad (1)$$

where k_1, k_2 and k_3 are, respectively, the splay, twist and bend elastic constants, $2\pi/q_0$ is the natural pitch of the material and χ is the susceptibility corresponding to the field $\mathbf{H}_{\text{eff}}$. One can then in principle find the director distribution $\mathbf{n}(x,y,z)$ that minimizes the free energy integrated over the specimen with the given constraints and boundary conditions.

Using a different approach, Abbate *et al.* [7] have shown that when

$$\frac{k_2 d}{\pi k_3}q_0>1 \quad (2)$$

where d is the cell thickness, the homeotropic alignment becomes unstable. The condition given by equation (2) defines the threshold for the changeover between the homeotropic and helical textures. To facilitate the discussion in the present paper we define the quantity ς as

$$\varsigma=\frac{k_2 d}{\pi k_3}q_0 \quad (3)$$

The threshold value for changeover between the homeotropic and the helical textures is defined by the

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condition

$$\varsigma = 1. \quad (4)$$

Since the elastic constants and the pitch are temperature-dependent, the parameter ς is expected to vary with temperature. In a material with the suitable temperature dependence of the elastic constants and the pitch ($2\pi/q_0$), one can cross the threshold by simply varying the temperature of a cell of fixed thickness d. Following the argument of Kamien and Selinger [3] one may expect that close to the threshold value there will be a region where solitons may be formed.

2. Experimental

The chiral nematic material used in the present work was prepared by doping a commercial room temperature multi-component nematic material (BLO32) with a small concentration (0.5–1 wt %) of chiral dopant C15. Both materials were obtained from Merck Ltd (UK) and were used without further purification. The mixture was heated to the isotropic phase with continuous stirring to ensure thorough and complete mixing; the mixture was then quickly cooled by quenching in ice. The process was repeated several times to ensure the formation of a homogeneous liquid crystal phase.

The cell was fabricated using glass plates treated with a solution of lecithin in chloroform (0.5 wt %). The cell gap was achieved by placing gauged Mylar spacers on the opposite sides of the cell. The cell was filled by capillarity at room temperature, and was hermetically sealed using an inert epoxy (NOA65 from Norland). Several cells with gaps in the range of 20–50 μm were prepared by this method.

The observations were carried out using a polarizing optical microscope in conjunction with a heating stage and temperature controller (Linkam TMS 94) with a temperature resolution of 0.1°C.

3. Observations

The parameter ς for the material prepared increases with temperature. A transition from the homeotropic alignment to the helical texture will be observed, provided that the cell thickness d is such that the temperature at which the threshold value of ς is reached is lower than the temperature of the transition to the isotropic phase. The observations presented in this section are divided into two sections: first the behaviour in cells of different thickness is presented, then the textures observed in a cell of constant thickness are described.

3.1. *Observations with varying cell thickness*

All the specimens prepared displayed a homeotropic alignment at room temperature and a helical structure at higher temperatures. As expected from equation (2) we observed that the temperature at which the textural change to the helical structure occurs decreases with increasing cell gap [7].

For small cell gaps the textural change does not occur uniformly at one critical value of ς. There are two critical values of ς: a lower value ς_1 below which the homeotropic alignment is stable and an upper value ς_2 at which the entire specimen switches uniformly to the helical texture. Between these two values an intermediate texture is observed. In this intermediate texture small circular domains with a helical structure are formed in the otherwise homeotropically aligned material. These circular domains observed only between the two critical values of ς are different from the 'bubble domains' previously reported in the literature [5]. The bubble domains are metastable domains that can exist far from the condition $\varsigma = 1$. Bubble domains were also observed in the present specimens. To avoid confusion hereafter, the intermediate texture observed between the two critical values of ς will be referred to as the '*bubble texture*'.

The temperatures at which the lower and upper critical values of ς are reached with different cells are given in the table. The uncertainty in the temperatures quoted in the table is (± 0.2°C). In the 50 μm thick cell the temperature range of the *bubble texture* is too narrow to measure. In thicker cells there is a direct change from the homeotropic to the helical structures. No homeotropic texture is observed when

$$d > \frac{\pi k_3}{k_2 q_0}.$$

3.2. *Observations in a cell of constant thickness*

All the observations reported in this section were made on the same specimen. The cell gap was 23 μm and the material contained 1 wt % of chiral dopant. The

Table. Temperatures at which the lower and upper critical values of the parameter ς are reached in cells of different thickness.

d/μm	T_{ς_1}/°C	T_{ς_2}/°C
23	86.9	88.7
36	86.7	88
50	87.1	87.5

sequence of textures observed on heating was

$$\text{Homeotropic} \xrightarrow{86.9^{\circ}C} \text{Bubble texture} \xrightarrow{88.7^{\circ}C} \text{Helical texture} \xrightarrow{91.5^{\circ}C} \text{Isotropic.}$$

It is emphasized that the texture changes are not driven by temperature but by the parameter ς. Cells of different thickness display the textural changes at different temperatures but the same value of ς.

At low ς, the texture is homeotropic and the twist is completely excluded from the specimen. At the lower critical values of ς, corresponding to 86.9°C, the first bubbles appear. Figure 1 (*a*) shows such bubbles viewed between crossed polarizers. The branches of the Maltese cross are approximately at 45° to the direction of the polarization of the incident light. When the bubble is just formed the centre of the Maltese cross is usually at the edge of the bubble. After a few seconds the centre of the cross evolves towards the centre of the bubble. Bubbles with the Maltese cross off-centred or at the edge can be seen in figure 1 (*b*). The diameter of the smallest bubbles observed is of the order of twice the cell thickness. If the temperature is held between the two critical values, fluctuations are observed; some bubbles disappear while new ones are formed.

As the temperature is increased, the density of bubbles increases. Some bubbles collide and merge to form larger bubbles. A merging event can be seen at the upper right-hand corner of figure 1 (*b*). The diameter of the bubbles varies continuously between 40 and 120 µm. Above a certain diameter the appearance of the bubble evolves: they become coloured, the brushes at the centre are narrower and they do not extend to the edges of the bubble. These large bubbles can be seen in figure 2 that was taken at a temperature just below the upper critical value of ς.

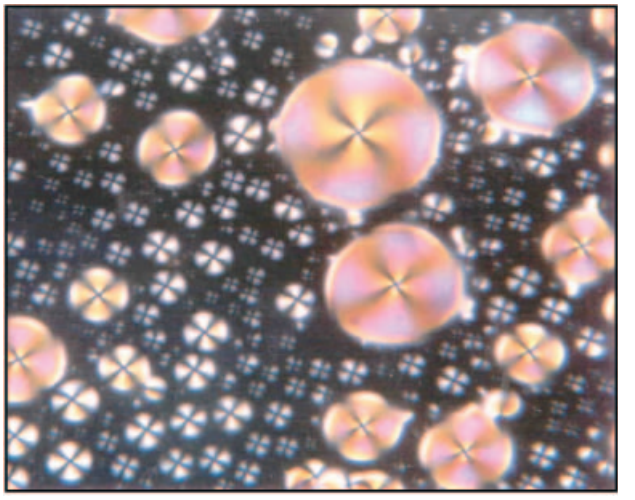

Figure 2. Bubble texture observed at values of the parameter ς just below the upper critical value.

At the upper critical value of ς corresponding to 88.7°C, the homeotropically aligned regions between the bubbles switch uniformly to the helical structure; the entire specimen adopts the texture show in figure 3. There are no defects marking the edges of the bubbles. This observation would suggest that in the bubble texture the director orientation is a continuous function across the boundary of the bubble. The singularities observed in figure 3 are the singularities that were at the centre of the large bubbles.

The textures thus described are not observed when cooling the specimen from the isotropic phase; in this case regular circular bubbles of cholesteric phase nucleate in the otherwise isotropic material. These bubbles merge to form large islands of cholesteric

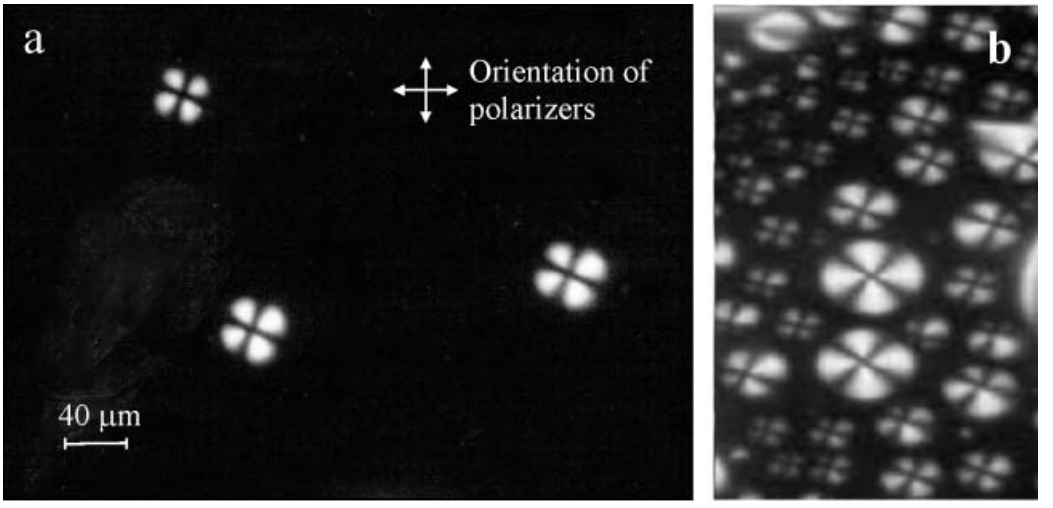

Figure 1. (*a*) Circular solitons observed at a value of ς just above ς_1. (*b*) At a temperature held between the two critical values, solitons continuously appear and vanish.

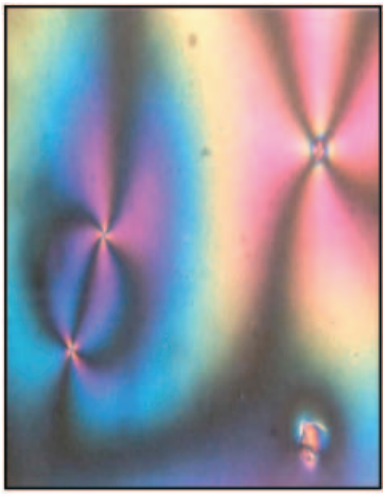

Figure 3. The specimen in the helical structure.

material. At lower temperatures, regions inside the islands become homeotropically aligned. On further cooling, the homeotropic texture extends to almost the entire specimen leaving only the previously reported fingers and loops with helical texture [5]. The boundaries of the parent bubbles become the walls of the fingers. These domains, fingers, loops and bubbles are metastable and supercool at room temperature far from the critical condition $\varsigma = 1$.

The walls of the supercooled domains are stable; they are not significantly affected if the specimen is heated to the temperature range where the whole specimen is in the helical texture. Figures 4 (*a*) and 4 (*b*), respectively, show these domains at temperatures where the homeotropic texture and the helical texture are favoured. The walls of the loop can clearly be seen as in-plane disclinations in the cholesteric material.

4. Discussion

The body of data presented in this paper does not permit an unequivocal description of the director distribution inside the bubbles. However, the following tentative argument could be made. As the parameter ς increases, the effective field decreases and reaches the threshold value first in the in the middle plane of the cell. The thickness of this middle plane may be a few molecular lengths and the effective field could be considered uniform across the plane. By minimizing equation (1) restricted to the middle plane, one expects as in Kamien and Selinger [3] to find a lower critical value of ς where the formation of solitons is possible. In their mathematical analysis Kamien and Selinger have constrained the helical axis to a uniform orientation. This constraint leads to the solution giving a soliton that is an infinite plane perpendicular to the helical axis. In the physical situation of the cell presented in this paper, it may be energetically more favourable for the soliton to bend and close back on itself instead of extending in an arbitrary direction across the entire cell.

The proposed interpretation presented in this paper for the formation of the bubbles is the following. Plane solitons as described by Kamien and Selinger nucleate in the middle plane of the specimen. As the length of the soliton increases, the two ends of the soliton attract each other causing the soliton to bend and close back on itself, thus forming the observed bubble. This tentative explanation, still to be confirmed, would be consistent with the observed evolution of the Maltese cross from the edge to the centre of the bubble.

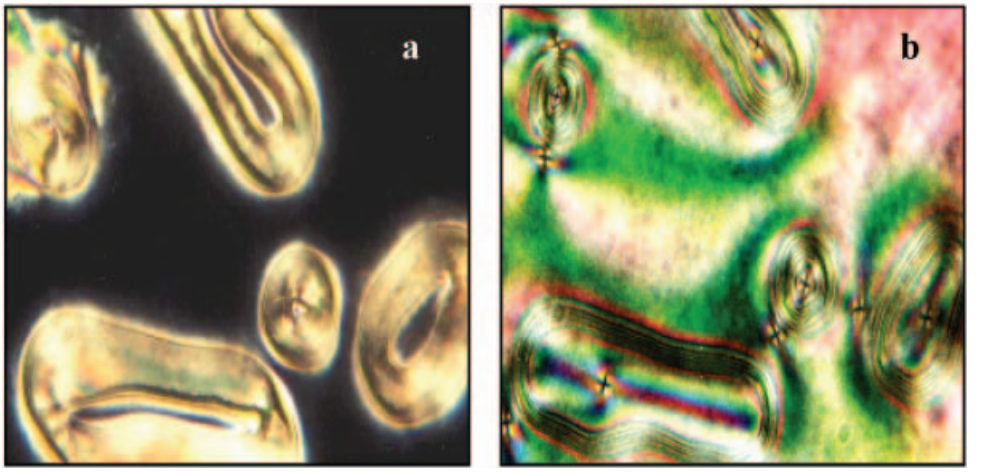

Figure 4. The metastable bubbles observed in (*a*) the homeotropic and (*b*) in the helical textures. In the helical structure there is a pair of singularities, one inside and one outside, associated with each bubble.

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