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Temperature induced anchoring transition in nematic liquid crystals with two-fold degenerate alignment

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The two-fold degenerate alignment of nematic liquid crystals provided by obliquely evaporated SiO_x layers was studied as a function of temperature. A temperature induced anchoring transition from tilted to planar alignment with more than 80 degrees between their preferred directions was found. The director in the two-fold degenerate region seems to follow a circular sector from the uniform tilted to the uniform planar anchoring, as the temperature increases. The anchoring transition is reversible and reveals the existing coupling between the azimuthal and polar angles of the preferred directions of alignment. The applicability of the two-fold degenerate alignment and related anchoring transitions of nematic liquid crystals for devices is briefly discussed.

1. Introduction

The orienting properties of obliquely evaporated silicon oxide on nematic liquid crystals was first reported by Janning [1]. SiO_x films evaporated at $\alpha = 85°$ to the substrate normal resulted in uniformly aligned liquid crystal layers with the director **n** lying parallel to the evaporation direction ($\mathbf{n} \| \sigma, 0 < \theta < 90°, \phi = 0°$), see figure 1. Shortly afterward, Guyon *et al.* [2] pointed out that the director does lie in the evaporation plane but with a larger pretilt from the substrate than first expected. They also found that the orientation of the liquid crystal director strongly depends on the parameters of the evaporation process, in particular the evaporation angle (see also [3, 4]). At a critical angle of incidence, which depends on the liquid crystal material used and the evaporation conditions, the director orientation changes from uniform tilted to uniform planar with the director perpendicular to the evaporation plane and with zero pretilt ($\mathbf{n} \perp \sigma, \theta = 90°, \phi = 90°$).

Subsequent studies [5–8] have shown that, in a narrow range of evaporation angles (usually in the range $62° \ \alpha \ 78°$), the preferred direction of the tilted anchoring splits up into two directions lying symmetrically with respect to the evaporation plane, see figures 1 (*b*) and 1 (*c*). This so-called two-fold degenerate tilted anchoring ($0 < \theta < 90°$, $0 < \phi < 90°$) may in turn result in twisted domains of both kinds of handedness in the cells [7, 9]. Moreover, the anchoring transition between the tilted and the planar anchoring is continuous and can occur both for small changes of the evaporation angle [5, 6, 8] and upon varying the thickness of the SiO_x layer [7, 10].

The first attempts to explain the alignment properties of obliquely evaporated SiO_x films involved studies of the topographic anisotropy of the film structures [11–13]. Berreman [14] used an elastic approach to describe the mechanism of liquid crystal alignment by solid surfaces. However, this elastic model requires the SiO_x layer roughness to be larger than the liquid crystal molecular length. In order to explain the anchoring on thin SiO_x layers and the observed continuous anchoring transition, Monkade *et al.* [7] suggested a model where so-called order electricity [15], due to the surface roughness of SiO_x, plays a major role. The alignment of nematic layers is determined by the anchoring of the liquid crystal molecules at the solid surface which may be affected by external factors, such as temperature, light and magnetic or electric fields. Indeed, the polar angle θ of the director, in the case of SiO_x aligning layers, appears to be temperature dependent [9, 16–19]. Likewise, strong electric fields can change the polar angle at the interface [20]. In the two-fold degenerate anchoring region, where both azimuthal and polar angles are of importance, it was found that these angles depend on the temperature [9]. Since two-fold anchoring of liquid crystal molecules has been suggested for applications [21], some emphasis is put in this paper on the question of the technical feasibility for this type of anchoring.

We study in particular the temperature dependence of the azimuthal angle of the preferred direction in nematic layers with two-fold degenerate alignment produced by

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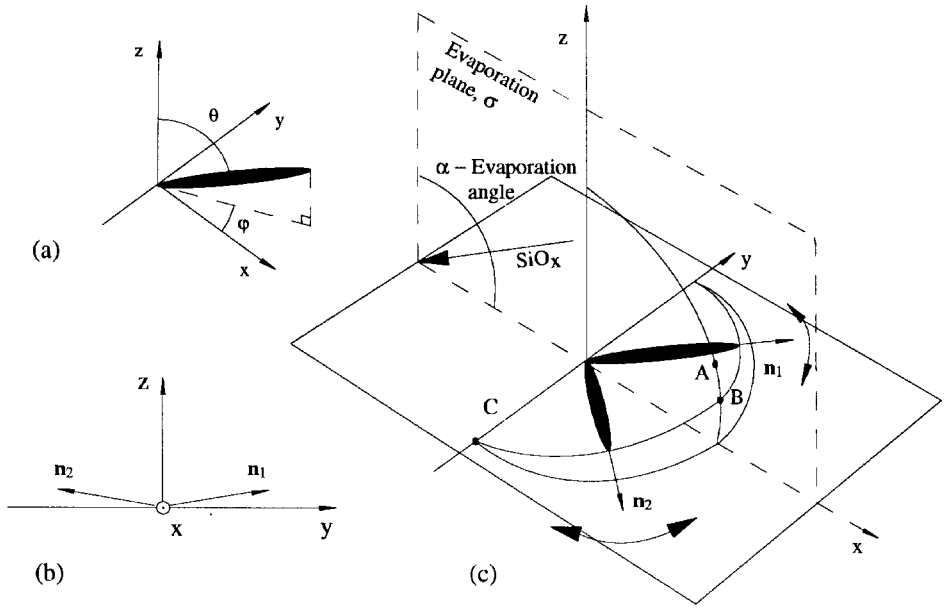


Figure 1. Schematic sketch of the anchoring transition in the region of two-fold degenerate alignment. The directions $\mathbf{n}_1$ and $\mathbf{n}_2$ represent the two possible director orientations at the liquid crystal – solid interface, and θ and ϕ the corresponding polar and azimuthal angles.

obliquely evaporated SiO_x and we discuss the observed anchoring transition.

2. Experimental

Our studies were performed using sandwich type cells with a cell gap of 6 μm. The glass substrates were ITO-coated soda lime float glass from Baltracon. The SiO_x deposition took place at room temperature and in high vacuum (10^{-7} mbar, SiO, Balzers BAK600). The set-up used for oblique evaporation was similar to the one originally described by Janning [1]. We have benefited from the natural spread of evaporation angles along the glass substrates to achieve cells with only slight changes in evaporation angle [5, 6]. The SiO_x thickness was measured by observing the frequency shift of a calibrated quartz oscillator. The SiO_x aligning layers were evaporated at 8 Å s^{-1} to a total thickness of 200 Å (perpendicular to the substrate plane). By assembling the glass substrates with their directions of evaporation antiparallel to each other, the most uniform alignment was achieved. Finally, the cells were filled in vacuum with the liquid crystal material in the isotropic phase and then cooled into the nematic phase. Four materials were used in our experiments: E7, ZLI-2806 (Merck), MBBA and 5CB (BDH). E7 and 5CB have positive dielectric anisotropy while ZLI-2806 and MBBA have negative dielectric anisotropy.

The observations of texture and anchoring transition were performed using a polarizing microscope with the liquid crystal cell kept in a hot stage (Mettler FP52), which controls the temperature of the cell during observation to within ±0·1°C. With crossed polarizer and analyser, it was straightforward to determine the extinction angles at various temperatures. Then, to conclude whether the molecules are parallel to the polarizer or to the analyser, a quartz wedge was used [22]. The measurements of the polar angle were performed according to the crystal rotation method [23].

3. Experimental results

3.1. *Two-fold degenerate tilted anchoring*

Two-fold degenerate tilted anchoring produced by obliquely evaporated SiO_x was found, in the case of E7, to exist in a narrower range of evaporation angles than reported before [5]. The measured values of the azimuthal angles ϕ as a function of evaporation angle α are plotted in figure 2, for SiO_x layers with thickness 200 Å. For the behaviour of the polar angle θ, we performed mostly qualitative measurements [24], see also figure 9 (*a*). Combining these results, the continuous transition of the director orientation between the uniform tilted anchoring and uniform planar, which takes place via the two-fold degenerate tilted anchoring, can be considered to have approximately the trajectory shown in figure 1 (*c*). Starting with SiO_x films evaporated at grazing incidence ($\alpha > 75°$), the director $\mathbf{n}$ lies in the evaporation plane with a pretilt larger than 20° (20° for $\alpha = 75°$) from the substrate and decreases as the evaporation angle decreases (from position A to position B, in figure 1 (*c*)). In this state the cells appear uniformly aligned and thus optically homogeneous. As the evaporation angle approaches 75°, the cells start to exhibit a 'sandy' texture. For 67° α 75°, a two-fold degenerate anchoring with symmetrically distributed anchoring directions, with respect to the evaporation plane, occurs. In fact, the two competing anchoring directions $\mathbf{n}_1$ and

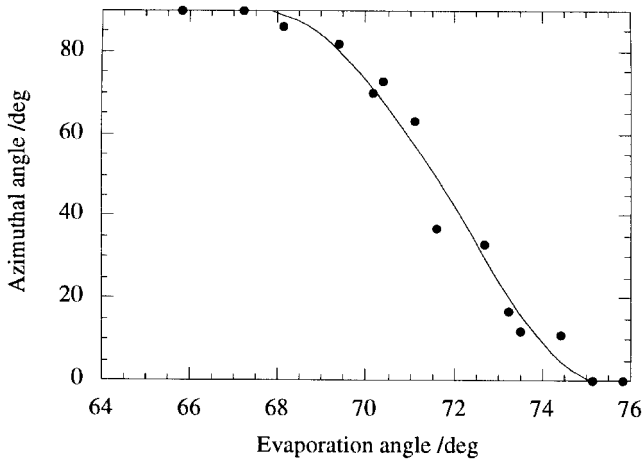


Figure 2. Azimuthal angle ϕ of the preferred direction of alignment in a cell filled with E7, as a function of evaporation angle α of the SiO_x layer with thickness 200 Å (The line is a fit to the experimental values).

$\mathbf{n}_2$ follow the intersection line of a sphere and a tilted plane perpendicular to the evaporation plane, through the region between B and C to position C in figure 1 (*c*) (see also [5, 7]). This indicates that there are two minima present in the anchoring energy of the nematic liquid crystal layer and hence two equivalent easy directions are available. The tilt angle (measured from the surface, $\psi = 90° - \theta$) continues to decrease continuously with decreasing α as the two anchoring directions approach the uniform planar anchoring. Finally, for cells evaporated with α 67° the nematic layer adopts a uniform planar anchoring, with the director perpendicular to the evaporation plane. Again, the cells appear optically homogeneous and show perfectly uniform alignment (position C in figure 1 (*c*)).

As a result, in the two-fold degenerate anchoring region, four types of domains can be distinguished in the texture of the liquid crystal layer, two with homogeneous alignment and two with twisted structures of opposite sense. In the domains with homogeneous alignment, the director is lying uniformly along one or the other of the two possible anchoring directions whereas in the twisted domains, the director has different directions at each cell surface corresponding to the two existing anchoring directions $\mathbf{n}_1$ and $\mathbf{n}_2$, respectively.

3.2. *Domains in the two-fold degenerate tilted region*

As mentioned above, four types of domains have been found in the cells with two-fold degenerate anchoring. Photographs of such a cell are shown in figures 3 and 4. Compared with figure 3, the sample in figure 4 has been rotated through an angle corresponding to twice the director azimuthal angle ϕ, which is the same as the angle between the two degenerate directions in the cell. The two types of domains that show extinction in figures 3 and 4, respectively, are the domains with alignment corresponding to one or other of the two symmetrically distributed directions of preferred alignment in the two-fold degenerate tilted anchoring. The other types of domains are the ones with twisted structure and they appear most often as a transition region between the two other types. The domains with twist do not show extinction for any position of the turn-table and the twist angle is equal to the angle between the two degenerate directions in the cell. As the evaporation angle decreases and the angle between the two degenerate directions increases, the corresponding twist angle also changes, as expected.

The selection of molecular direction in each domain seems to occur accidentally throughout the entire sample and in most cells, domains with the two degenerate anchorings are mediated by domains with twist structure. Jérome *et al.* pointed out that by using a controlled wetting process [6, 25], the different anchoring directions can be selected during filling. The cells maintain the same domain-pattern even if they are heated up above the nematic–isotropic transition temperature and then cooled down again. The selection of preferred alignment direction is probably established during the filling and first cooling process and then preserved.

The size of the domains seems to depend strongly on the evaporation angle α. Within the range of the two-fold degenerate tilted anchoring, the higher the evaporation angle the larger are the domains observed in the cells. This is illustrated in figures 5, 6 and 7, where the domain pattern of three cells, with SiO_x aligning layers evaporated at different angles of incidence, are shown. It is likely that the size of the domains is affected by the structure of the underlying SiO_x, showing great dependency on the evaporation angle. In some cells just one single or a few domains were completely dominant, these cells otherwise showing the same behaviour as the other cells.

3.3. *Temperature induced anchoring transition in the two-fold degenerate alignment region*

As we have seen, in cells with two-fold degenerate tilted anchoring, the director azimuthal and polar angles, both depend on the evaporation angle. Moreover, there is a coupling between them (cf. [5, 7]) and, as mentioned in the Introduction, the polar angle is temperature dependent (see also figure 9 (*a*)). Consequently, we may also expect the azimuthal angle to be temperature dependent. Indeed, in this study we observed that in cells with two-fold degenerate anchoring, the values of the azimuthal and polar angles can both be controlled thermally. The effect of increasing temperature on the azimuthal angle is the same as that for decreasing the

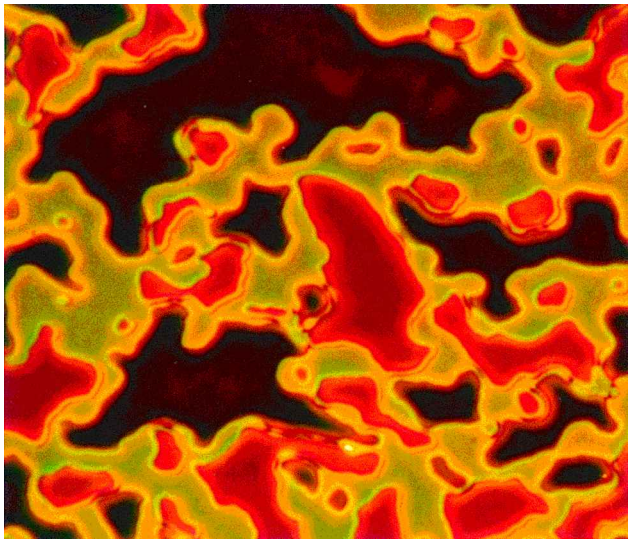


Figure 3.

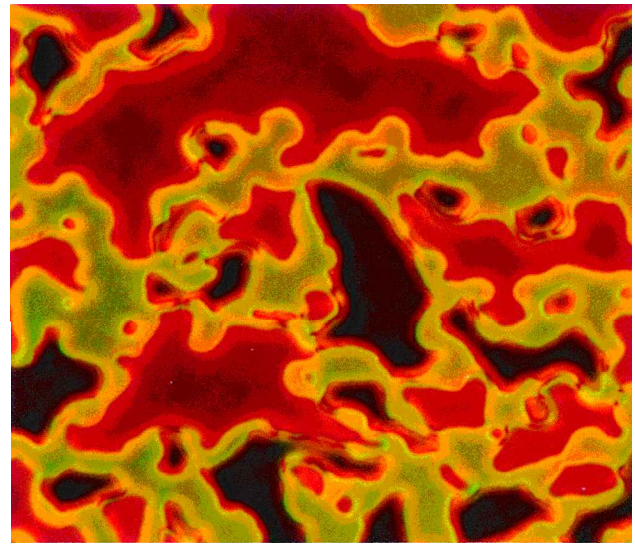


Figure 4.

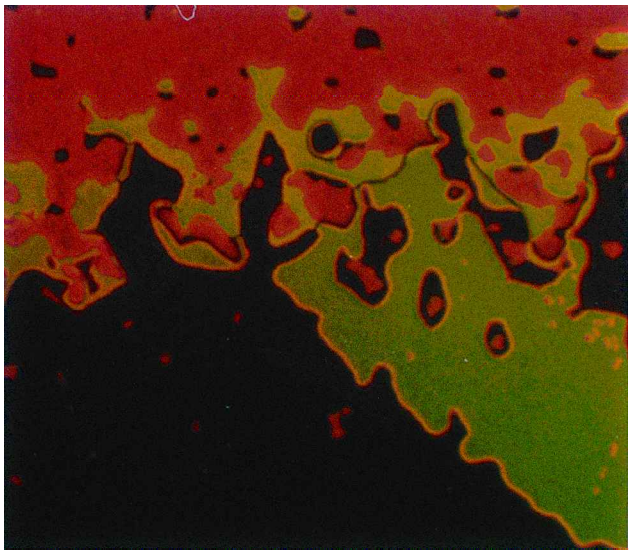


Figure 5.

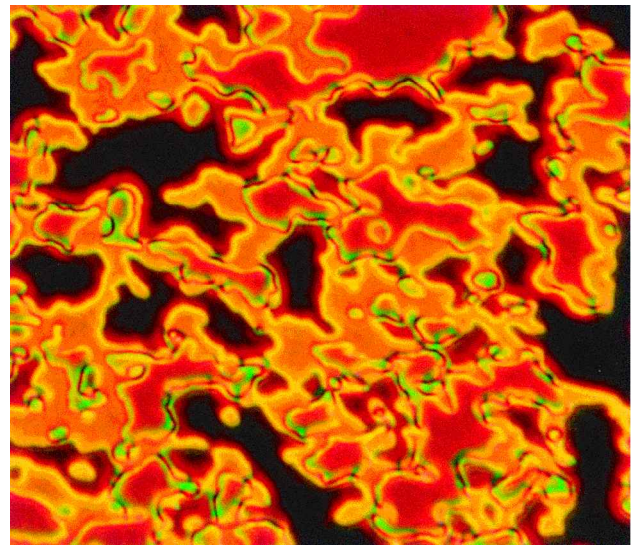


Figure 6.

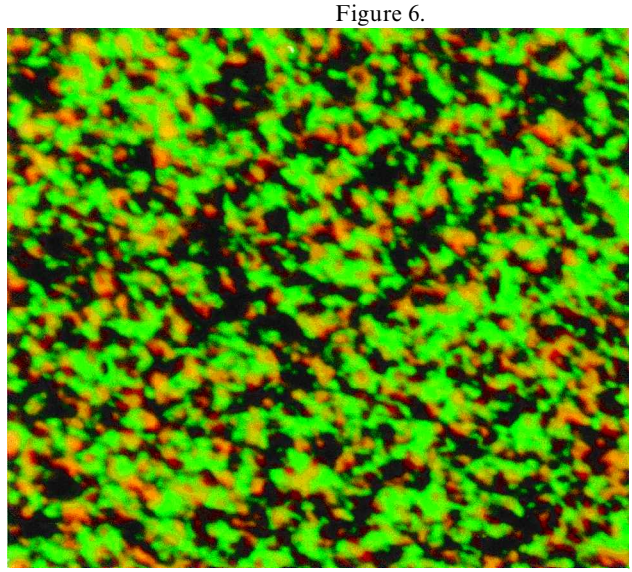


Figure 7.

evaporation angle, namely that the preferred directions of alignment of the liquid crystal molecules approach the uniform planar orientation perpendicular to the evaporation plane. The preferred direction of alignment changes from the initial position, somewhere between positions B and C in figure 1 (*c*), towards position C. Domains initially having a large value of the azimuthal angle (the case of low evaporation angles) exhibit a small increase of the azimuthal angle when increasing the temperature, whereas in domains with initially small azimuthal angles (high evaporation angles) the change is larger. This is demonstrated in figure 8 where the temperature induced changes in the values of the azimuthal angle are plotted for the cells with two-fold degenerate anchoring for evaporation angles in the same range as in figure 2 ($67° \alpha 75°$). The common tendency in all cells is that the azimuthal angle of the preferred direction of alignment increases with temperature until the alignment becomes uniform planar, i.e. the azimuthal angle reaches $\sim 90°$.

It was found that the two symmetrically distributed anchoring directions change equally with temperature. As a consequence, when the temperature increases and the two-fold degenerate alignment transforms into a uniform planar alignment, perpendicular to the evaporation plane, the entire sample shows extinction for the same position of the turn-table in orthoscopic studies. In other words, the two-fold (bistable) anchoring transforms into a uniform (monostable) anchoring.

As expected, the magnitude of the pitch in the twisted domains changes when the temperature increases, since the ends of the helix in the twisted structure follow the two degenerate directions, and at high temperatures the twist angle reaches zero. Hence, the twisted domains show full light extinction under the same conditions (temperature, polarizer settings) as the other domains.

During measurement of the azimuthal angle as a function of temperature, in the two-fold degenerate anchoring region, we allow the sample to stabilize at each chosen temperature. Yet, there is a hysterisis between the heating and cooling runs. The difference in measured values of the azimuthal angle is up to between 10 and 20 degrees of arc for the same temperature. Moreover, the rate of change of azimuthal angle with temperature varies strongly from cell to cell, cf. figures 9 (*a*), 9 (*b*) and 9 (*c*). It occurs very rapidly at a threshold temperature in the cells with high evaporation angle, while in cells with low evaporation angle, the change has a more linear character. It can also be seen that the threshold temperature varies with evaporation angle. A high evaporation angle results in a high onset temperature for the beginning of the anchoring transition, as shown in figure 10. It seems that the change of the preferred direction of alignment near the uniform tilted anchoring brings about larger changes in the director field than does the corresponding one near the uniform planar anchoring. It is likely that this difference depends on the structure of the evaporated SiO_x. Therefore, cells with a thickness of the SiO_x layer ranging from 120 to 280 Å were produced and investigated. However, no significant change in the character of the anchoring transition occurred.

Just as in the case of E7, domains with two-fold degenerate anchoring were found in cells filled with two other materials, MBBA and 5CB. The limits in the evaporation angle of SiO_x for achieving the three different types of alignment (tilted, two-fold degenerate and planar) that were observed for the cells with E7, are shifted towards higher evaporation angles. The temperature induced anchoring transition was also found in these cases, being more pronounced in cells with MBBA than with 5CB. A quite different behaviour was found in cells filled with the nematic material ZLI-2608, for which the transition from the uniform tilted to the

Figure 3. Domain pattern in a cell with two-fold degenerate alignment produced by obliquely evaporated SiO_x. The cell is filled with E7 and viewed between crossed polarizers (333 ×).

Figure 4. The same sample as in figure 3 but rotated through 20°. Here the domains with the other of the two degenerate anchoring directions are in the optical extinction position. The domains with twist do not show extinction for any position of the turn-table.

Figure 5. A cell with SiO_x aligning layer evaporated at $\alpha = 74°$. The green areas are the domains with twisted structure and the red and the dark areas are the domains possessing one or other of the two-fold alignment directions. The size of the domains is larger than in cells with SiO_x layers evaporated at lower angles (333 ×).

Figure 6. A cell with SiO_x aligning layer evaporated at $\alpha = 73°$. The yellow areas are the regions showing twisted structure and the red–brown and the dark areas are the domains with one of the two degenerate anchorings. The domains are 'middle-sized' (333 ×).

Figure 7. A cell with SiO_x aligning layer evaporated at $\alpha = 70°$. The green areas are the regions showing twisted structure and the yellow and the dark areas are the domains with one of the two degenerate anchorings. The domains are relatively small compared with those in figures 5 and 6 (333 ×).

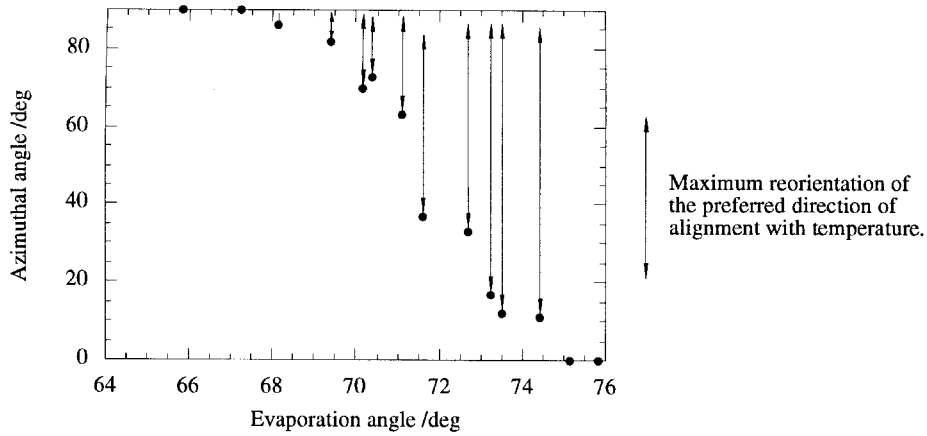


Figure 8. Temperature induced change of the azimuthal angle of the preferred direction of alignment in cells with two-fold degenerate anchoring as a function of the evaporation angle α. The lines represent the maximum magnitude of these changes when the temperature varies from T_{room} to T_{iso}; the cells are filled with E7.

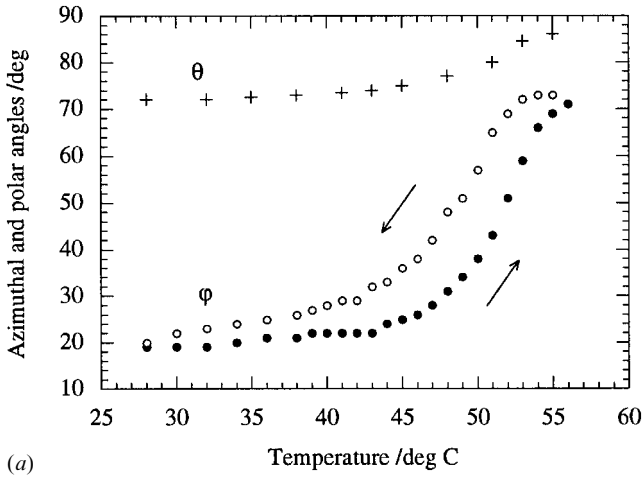
uniform planar anchoring is very sharp. When heating the sample, the two-fold degenerate anchoring can be seen as a narrow transition region which separates the areas with tilted and planar anchoring. The small temperature gradient that naturally occurs inside the sample on heating is thus enough to allow co-existence of all three types of alignment. However, despite the differences between the liquid crystal materials, the general behaviour with respect to director trajectory shown in figure 1 (*c*) is the same for all tested materials. In all investigated cells, the temperature induced transition has a reversible character with a hysterisis as described for E7 above.

4. Discussion

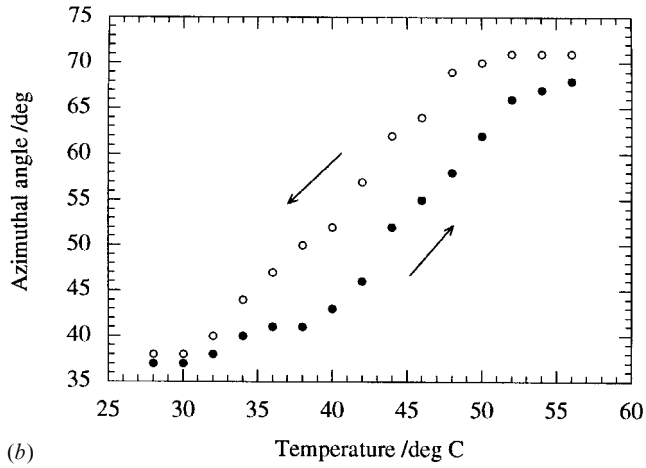
During the last few years, much attention has been given to bistable switching in nematics due to its potential for applications. This type of switching, however, is not an intrinsic feature of nematic materials as it is for ferroelectric smectics [26]. The bistable switching in nematics can be obtained only in the presence of a two-fold degenerate anchoring produced by special surface treatments [27]. The achievement of such anchoring is not an easy task because of its sensitivity to the choice of a number of technological parameters which in turn depend on the type of liquid crystal material used. Moreover, it is difficult to obtain a uniform alignment over the whole cell area under two-fold degenerate anchoring conditions. Our present experimental study shows that only in a limited number of cells was one single or just a few domains with one of the two-fold tilted anchorings found, while in the majority of cells, the entire area was covered with domains of varying sizes and directions of preferred alignment. As described in § 3·2, it was in cells with high evaporation angles (within the two-fold alignment) that we were able to obtain single domains. It seems that the influence of wetting is not strong enough to prevent accidental selection of anchoring in the other cells.

Previous studies of two-fold degenerate anchoring have shown that the polar and azimuthal angles are coupled and that their magnitude can be controlled by, for instance, the evaporation angle. It is likely that the preferred direction of alignment follows the trajectory shown in figure 1 (*c*), where its initial position is defined by the evaporation angle. The pretilt (from the substrate) of the director of nematics aligned by SiO_x layers has been reported to decrease with increasing temperature, as described above. The pretilt in our cells exhibits the same behaviour (see figure 9 (*a*)). Moreover, we also found that the azimuthal angle is temperature dependent, thus revealing once more the coupling between the polar and the azimuthal angle. Our experiments show that the azimuthal angle increases while the pretilt decreases on increasing the temperature, a behaviour similar to that observed on decreasing the evaporation angle in the region of the two-fold degenerate anchoring. The temperature dependence of the azimuthal angle means that the preferred directions of the two-fold degenerate alignment can be changed by varying the temperature. In fact, the observed temperature induced anchoring transition exhibits just the opposite character to the one reported in [9].

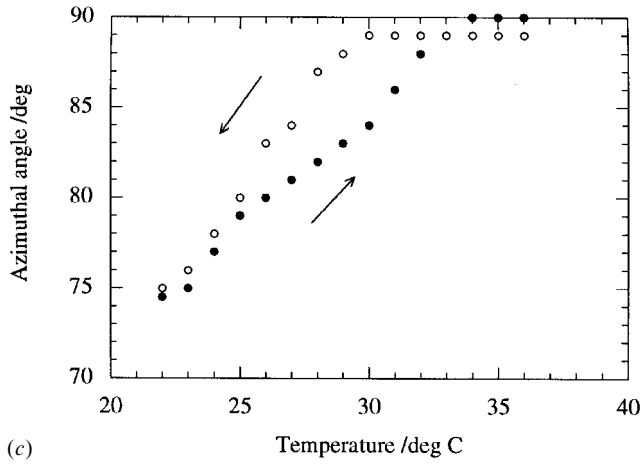
The coupling existing between the two angles was recently unambiguously demonstrated by applying an electric field across the cell [28]. If the field strength is sufficient to change the pretilt angle of the liquid crystal molecules near the substrate, through the dielectric coupling, then the azimuthal angle also changes; this results in a change of position of the preferred direction of alignment in the plane of the sample, similarly to the temperature driven anchoring transition. In addition, the azimuthal angle of the preferred direction of align-



(a)



(b)



(c)

Figure 9. Temperature induced change of the preferred direction of alignment in the region of two-fold anchoring for cells with different evaporation angles: (*a*) $\alpha = 73{\cdot}5°$, (*b*) $\alpha = 72{\cdot}5°$ and (*c*) $\alpha = 70{\cdot}5°$. Plot (*a*) contains measurements of both the azimuthal and polar angles; in (*b*) the change has a more linear character than in (*a*); in (*c*) only a small change takes place and begins already at room temperature. The cells are filled with E7.

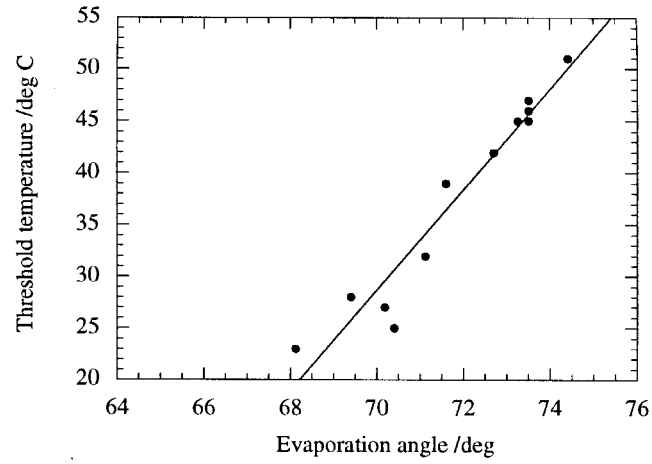


Figure 10. Threshold temperature for the temperature induced anchoring transition. The line is a linear fit to the experimental values; material: E7.

ment can be affected by applying an in-plane electric field, thus resulting in a change of the pretilt angle.

In the two-fold degenerate anchoring region, the equilibrium between forces of an elastic and a chemical nature results in two local minima defining the position of the directions of preferred alignment. This equilibrium can easily be affected by temperature or by external fields, as the experiments have shown, thus resulting in a change of the directions of preferred alignment. The coupling between the azimuthal and polar angles demonstrates that the common artificial division of the anchoring energy into azimuthal and polar parts is an oversimplification, and that we must consider the interactions between liquid crystals and solid substrates in three dimensions. A theoretical model of the observed anchoring transitions in the two-fold anchoring region induced by temperature, as well as by an external field, will be published elsewhere [29]. In order to obtain a better understanding of the character and the potential of possible applications of these transitions, further studies with different nematic materials and surface treatments are required. For applications, however, the surface treatment has to result in uniform cells with only one type of domain that can be switched either by temperature or by electric fields.

5. Conclusions

We have studied a temperature induced anchoring transition in nematic liquid crystals with two-fold degenerate alignment, produced by oblique evaporation of SiO_x ($67° \ \alpha \ 75°$). During the transition, the preferred direction of alignment seems to follow a circular sector between uniform tilted anchoring, in the evaporation plane, and uniform planar anchoring, perpendicular to the evaporation plane. The transition, which is symmet-

ric with respect to the evaporation plane for both easy directions in two-fold degenerate anchoring, reveals the coupling between the azimuthal and polar angles. During the temperature driven anchoring transition, the azimuthal angle of the preferred directions of alignment increases on increasing the temperature. This transition was studied for the four nematic liquid crystal materials E7, MBBA, 5CB and ZLI-2608. It was found to be reversible and to be most pronounced in the material E7, with a maximum of the temperature induced change in the azimuthal angle of more than 80 degrees of arc. Moreover, the transition exhibits a non-linear behaviour and occurs at some threshold temperature which in turn depends on the evaporation angle. The temperature driven anchoring transition in the region of the two-fold degenerate anchoring raises the question whether this type of anchoring is convenient or not for use in liquid crystal displays, where temperature independent characteristics are desirable. The observed temperature dependence certainly sets some practical limits for the applicability of this kind of alignment in bistable nematic electro-optic devices. However, the coupling between the azimuthal and polar angles may be of practical use. A local heating, by a laser beam or otherwise, can effectively change the preferred directions of alignment. Thus, a 'light controls light' liquid crystal device may be based on this effect [30]. Possibly, the hysteretic character of the temperature induced anchoring transition may also be of practical use.

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