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OPTICALLY ACTIVE ADDITIVES IN TWISTED NEMATIC DEVICES (*)

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Résumé. — Une étude a été faite concernant l'emploi d'additifs optiquement actifs dans des dispositifs formés de nématiques tordus, afin d'éliminer les domaines de torsion inverse. Les résultats dépendent de la chiralité des additifs et de l'alignement des directeurs sur les deux parois. Sous certaines conditions, la dégénérescence de réorientation du directeur induite par le champ fait apparaître des taches, même en l'absence de torsion inverse. On montre comment on peut lever cette dégénérescence pour obtenir une bonne uniformité de présentation.

Pour des combinaisons particulières de chiralité et d'alignement de surface, on montre que l'application d'un champ électrique suffisamment fort produit un renversement temporaire de la chiralité de la couche.

Un calcul du seuil de réorientation de couches nématiques tordues contenant des additifs optiquement actifs est comparé à l'expérience.

Abstract. — A study has been made of the use of optically active additives in twisted nematic devices to remove areas of reverse twist, the results obtained depending on the chirality of the additives and the director alignment on the two containing surfaces. Under certain conditions degeneracy of the field induced reorientation of the director produces a patched appearance even in the absence of reverse twist. It is shown how to remove the degeneracy to obtain good display uniformity.

For particular combinations of chirality and surface alignment it is shown that the application of a sufficiently strong electric field produces a temporary reversal of the chirality of the layer.

A calculation of the threshold condition for the field-induced reorientation of twisted nematic layers containing optically active additives is compared with experiment.

1. Introduction. — The performance of twisted nematic electro-optic devices [1] is often spoiled by the occurrence of patches of non-uniform contrast. Some of these patches are caused by areas of reverse twist [2], and can be eliminated by using long pitch cholesteric materials made by incorporating optically active additives in nematics [3]. We have therefore examined the reorientation by an electric field of $\pi/2$ twisted layers of long pitch cholesterics.

2. Materials. — Cholesterics with pitch $P = 80 \mu\text{m}$ can be used in making $\pi/2$ twist devices of thickness $d \leq 40 \mu\text{m}$ [3]. Such materials were made by adding to the nematic 4'-n-pentyl-4-cyanobiphenyl (5CB) [4] either 0.2 % by weight of cholesteryl nonanoate (CN) or 2 % by weight of 4'-(2"-methylbutyloxy)-4-cyanobiphenyl (50CB) [5]. In the experiments which required a nematic material, pure 5CB was used.

3. Reverse tilt. — Some devices with a $\pi/2$ twist incorporating these long pitch cholesteric materials still exhibited patches when an electric field was applied.

These patches were separated by lines seen as broad and diffuse in the microscope (Fig. 1), and are caused by areas of reverse tilt. If the tilt angle of the director in the middle of the layer, ϕ_M , is zero, then in the steady state the field will produce either clockwise or anti-clockwise reorientation about an axis normal to both the field and the director. This degeneracy of reorienta-

Fig. 1. — Broad field-dependent disclination lines separating areas of opposite tilt.

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tion was first observed by Williams [6] and correctly interpreted by Helfrich [7]. There can be a non zero tilt angle at both containing surfaces (Fig. 2), and for

FIG. 2. — Definition of tilt angles of the director at the surfaces and middle of a nematic layer.

simplicity we first examined the common combinations for nematic layers with a zero twist angle. The results are summarized in table I in which we give the values

TABLE I

Occurrence of reverse tilt for various common combinations of surface tilt angles for zero twist nematic layers

	Tilt angles at the two surfaces	Tilt angle at the middle (ϕ_M)	Reverse tilt ?
1	$\phi_1 = 0, \phi_2 = 0$	0	Yes
2	$\phi_1 \neq 0, \phi_2 = 0$	$(\phi_1/2)$	No
3	$\phi_1 \neq 0, \phi_2 \neq 0$		
(a)	$\phi_1 = \phi_2$	ϕ_1	No
(b)	$\phi_1 = -\phi_2$	0	Yes

of ϕ_M and the incidence of patches of reverse tilt. Case 3 is the most complex one, and figure 3 illustrates the two possibilities. The layer with $\phi_1 = \phi_2$ has a uniform tilt

FIG. 3. — The two possible configurations in zero twist layers with $\phi_1 \neq 0$ and $\phi_2 \neq 0$. a) *Uniform* (uniform tilt angle). b) *Splayed* (varying tilt angle).

angle and is called a *uniform* layer. If $\phi_1 = -\phi_2$, the tilt angle varies across the layer which is described as *splayed*. If nematic layers with a $\pi/2$ twist angle are now considered, then the results of cases 1 and 2 of table I were found to be applicable. However case 3 was more complex, and for a particular combination of ϕ_1 and ϕ_2 , a $\pi/2$ twisted layer was either splayed or uniform depending on the sense of twist of the layer. Models of such layers make this fact self-evident. It is demonstrated by figure 4, in which patches are seen in only one sense of twist in a nematic layer with a $\pi/2$ twist angle and both ϕ_1 and $\phi_2 \neq 0$.

FIG. 4. — Nematic layer with both $+\pi/2$ and $-\pi/2$ twist angles showing reverse tilt in only one sense of twist angle.

The observations made when an electric field was applied to a long pitch cholesteric layer with a $\pi/2$ twist angle are a simple extension of the above if it is remembered that the optically active additive has the effect of making one sense of twist energetically favourable. The twist of the cholesteric therefore interacts with the asymmetry of the alignment on the two surfaces. Figure 5 shows the plan view of the alignment necessary for producing a uniform layer if either

FIG. 5. — Plan view of surface alignment necessary for producing uniform $\pi/2$ twisted layers of long pitch cholesterics using cholesteryl esters or 50CB. The arrow indicates the direction in which the director tilts out at the surface.

cholesteryl esters or ^{*}50CB are used as the additive. If the cholesteric has the opposite sense of twist (eg by using Canada balsam as the additive), then the opposite combination of alignment to that in figure 5 is necessary to produce a uniform layer.

4. Patch-free twisted nematic devices. — For good device appearance patches of reverse twist and reverse tilt have to be eliminated. The former are removed by using the long pitch cholesterics already described, and the latter by using the correct combination of φ_1 and φ_2 .

These are :

- a) $\varphi_1 = 0$ and $\varphi_2 \neq 0$, together with any sense of cholesteric twist ;
- b) $\varphi_1 \neq 0$ and $\varphi_2 \neq 0$, together with the correct sense of cholesteric twist.

Various techniques exist for producing alignment on a surface, and table II shows the values of φ (for 5 CB) for these techniques that we have measured using the conoscopic method.

TABLE II

Tilt angles for 5CB on glass surfaces using various alignment techniques

φ	Alignment technique
0°	Evaporation of SiO ₂ , etc. at 30° grazing angle
≈ 25°	Evaporation of SiO ₂ , etc. at 5° grazing angle
≈ 2°	Unidirectional rubbing of a clean surface or surface coated with surfactant
≈ 2°	Unidirectional rubbing along the alignment direction of a surface with $\varphi = 0^\circ$

5. Transient reorientation phenomena. — We have observed two transient phenomena which are a consequence of various angles of tilt. When a moderate voltage (eg 10 V rms) was applied to splayed layers with either zero or $\pi/2$ twist angles, we observed a transient reorientation which was coloured between crossed polarizers and which reverted to the usual steady state reorientation after one or two seconds. An explanation of this effect can be found by reference to the illustration of a splayed layer in figure 3b. In the usual steady state reorientation the middle of the layer deforms first. However it is also possible for the middle of the layer to remain unchanged with $\varphi_M = 0$, whilst the top half of the layer reorients clockwise and the bottom half anticlockwise. This reorientation would have higher elastic and electrical energy than the usual reorientation and would therefore be unstable.

A second transient phenomenon was observed during the fast switching of a twisted nematic device [8] using the frequency dependent dielectric anisotropy of the Merck W 1 material. Layers with an initial $\varphi_M \neq 0$ were used so that there were no reverse tilt patches when the low frequency signal was applied. However we observed that if the high frequency signal was applied for ≥ 10 mS just prior to the application of the low frequency, then a considerable number of reverse tilt patches was produced. This is explained by the fact that the high frequency signal forced the majority of the layer to adopt a zero tilt angle.

6. Electric field induced change of twist angle of splayed layers. — When a moderate voltage (eg 10 V rms) was applied to a splayed layer for a few seconds, a change of twist angle occurred, and if the field was then removed the layer had become uniform. The change of twist angle was confirmed by observing the orientation of the Williams' domains in suitable materials (see for example reference [2]).

Examples of this change of twist angle are that a splayed $\pi/2$ layer was converted into a uniform $-\pi/2$ layer, and secondly a splayed zero twist layer was converted into a uniform π layer. The effect in a $\pi/2$ twisted layer was strong enough to overcome the effect of an optically active additive.

To confirm that the transition was only a change of twist angle a wedge cell was constructed (Fig. 6) using

FIG. 6. — Wedge cell to demonstrate the change of twist angle

the long pitch cholesteric. Because of the varying twist angles (0, π , 2π , etc.) the wedge contained alternate uniform and splayed layers. The dividing disclination lines moved when a field was applied, with all the uniform areas growing at the expense of the splayed areas, until eventually they covered the whole wedge. Removal of the field left a wedge with areas with twist angles 0, 2π , 4π , etc.; the original twist angles returned after several hours. This phenomenon had already been observed by Scheffer [9]. The field-induced change of twist angle is caused by the higher energy of the deformed splayed layer compared to the uniform layer. Both of these are illustrated in figure 7, and it seems likely that the splayed layer has a higher elastic and electrical energy. This excess energy must be greater than that of the unfavourable twist for the change of twist angle to occur.

FIG. 7. — Deformed splayed layer being transformed into the lower energy deformed uniform layer (the change of twist angle is not shown).

7. Threshold voltage for reorientation of $\pi/2$ twisted long pitch cholesteric layers. — The optically active additive is expected to slightly increase the threshold for reorientation by an electric field. Using the continuum theory we have calculated the threshold voltage (V_{cc}) for $\pi/2$ twisted cholesteric layers. V_{cc} is related to the threshold voltage (V_{cn}) for the equivalent layer of pure nematic by :

$$V_{cc}^2 = V_{cn}^2 + \left(\frac{2d}{P}\right) \left(\frac{\pi^2 k_{22}}{\epsilon_0 \epsilon_a}\right)$$

where V_{cn} has the usual form :

$$V_{cn}^2 = \frac{1}{\epsilon_0 \epsilon_a} \left[k_{11} \pi^2 + (k_{33} - 2k_{22}) \left(\frac{\pi}{2}\right)^2 \right]$$

k_{11} , k_{22} and k_{33} are the Frank elastic constants, and ϵ_a the dielectric anisotropy. The small quantities of additives used mean that these constants will be equal to those of the pure nematic. Berreman [10] has recently obtained the same expression for V_{cc} . It is interesting to note that the effect of the optically active additive is simply to add an extra term onto V_{cn}^2 proportional to the quantity $(\pi^2 k_{22}/\epsilon_0 \epsilon_a)$. This quantity can easily be determined from the cholesteric to nematic phase change in mixtures containing a few percent of CN in the nematic, and the value for 5 CB is 0.43 V^2 [11].

V_{cc} for various values of $(2d/P)$ was measured using

a polarizing microscope to observe a wedge with thickness d varying from $6 \mu\text{m}$ to $75 \mu\text{m}$. It is well known that the optically observed threshold is higher than the true threshold [12], so we examined the nematic 5 CB in an identical wedge. Figure 8 shows the values of the threshold voltage of both the cholesteric and the pure nematic measured optically. The difference between the two should be close to the true difference ($V_{cc}-V_{cn}$) caused by the optically active additive. Figure 8 shows this measured difference and

FIG. 8. — Threshold voltage for reorientation of nematic and long pitch cholesteric layers measured optically. The experimental difference caused by the optically active additive is compared with the calculated difference.

the calculated difference. The discrepancies between these is possibly mainly attributable to the optical method, and more accurate determinations of V_{cc} and V_{cn} using the capacitance method are now being carried out.

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