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Volume-stabilized ULH structure for the flexoelectro-optic effect and the phase-shift effect in cholesterics

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Flexoelectric coupling gives rise to a linear electro-optic response in cholesterics (flexoelectro-optic effect) with a uniformly lying helix (ULH) structure and this electro-optic effect is strongly reliant on the homogeneity and quality of the texture. The ULH structure, unfortunately, is complicated in itself and may be perturbed by factors such as dielectric coupling, surface/liquid crystal interactions and phase transitions, and often there is a tendency for relaxation into the Grandjean texture (standing helix structure) with time. Hence, in order to exploit the flexoelectro-optic effect in cholesterics any instability of the ULH structure must be ruled out.

We have overcome these problems by incorporating a polymer network by means of photopolymerization of a reactive monomer added to the cholesteric. The volume stabilized ULH structure still exhibits the flexoelectro-optic effect, it is stable and it is also retained after heating to the isotropic phase and going back to the cholesteric phase. In addition to the flexoelectro-optic mode, the ULH structure is of interest in an electro-optic mode characterized by a pure phase-shift with no change in amplitude (transmittance). This mode, which has obvious applications in spatial light modulators, optical computing devices and electrically controlled kinoforms and phase holograms working without polarizers, is also briefly discussed.

1. Introduction

The flexoelectro-optic effect in cholesteric liquid crystals was first described by Patel and Meyer in 1987 [1]. The macroscopic optic axis of a uniformly lying helix (ULH) cholesteric swings out (tilts) in the plane of the cell under the action of an electric field applied normal to the cell glass plates. The field-induced tilt of the optic axis is found to be a linear function of the field for a quite unusually large range of applied field E according to

$$\phi \approx \frac{e}{Kk} E \tag{1}$$

where

$$e = 1/2(e_s + e_b) \tag{2}$$

and

$$K = 1/2(K_{11} + K_{33}) \tag{3}$$

are the average values of the splay and bend flexoelectric coefficients and elastic constants, respectively, and k is the helical wave vector [2]. The sign convention used for flexoelectric coefficients agrees with that of Schmidt *et al.* [3] and also with Patel and Meyer [1], though not with the original Meyer paper [4], and is discussed at length in Rudquist and Lagerwall [5].

A severe problem in the flexoelectro-optic effect is that the ULH structure is unstable. It can be irreversibly damaged by external factors such as dielectric coupling, the influence from the surfaces of the cell, and phase transitions. Negative dielectric anisotropy $\Delta\varepsilon = \varepsilon_\parallel - \varepsilon_\perp$ tends to make the ULH structure transform into a standing helix (Grandjean) texture when applying the electric field. Thus a positive dielectric anisotropy is preferred. However, if the magnitude of $\Delta\varepsilon$ is large, the field-induced unwinding of the helix due to dielectric coupling strongly distorts the electro-optic response and may even destroy the ULH alignment. Hence, $\Delta\varepsilon$ should be positive and very close to zero, but as the dielectric anisotropy is often dependent on both temperature and frequency, its variation, as well as the possibility even of a change in sign, must be taken into account [6, 7].

Since neither planar nor homeotropic surface conditions fully support the ULH structure, degradation of the alignment occurs. The structure is also generally destroyed by phase transitions, i.e. the ULH texture is not retained after heating (or cooling) to another phase and then going back to the cholesteric phase. Any

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commercial device must withstand large temperature variations and work properly even after being subjected to extreme temperature conditions, for instance during storage and transport.

In this work we present a way to stabilize the uniformly lying helix structure by incorporating a polymer network in the volume of the ULH cell and we discuss the influence of the network created on the electro-optic characteristics of the flexoelectro-optic effect. We also suggest another application of the polymer stabilized cholesteric ULH structure working in dielectric mode, where the field-induced unwinding of the helix continuously changes the macroscopic birefringence of the cell. Thus we have two electro-optic effects suitable for the volume stabilized ULH structure. First, the flexoelectro-optic effect, in which e should be large and $\Delta\varepsilon$ small, and second, the unwinding effect in which the dielectric anisotropy should be large and e small.

2. Experimental

A cholesteric liquid crystal mixture TI827, kindly supplied by Merck Ltd, was used in our experiments as a material exhibiting a large flexoelectric tilt of the optic axis. It has a short pitch (0·3 μm), constant over a wide temperature range (≈0–50°C), and a low positive dielectric anisotropy. The liquid crystal material was mixed with 10 wt % of a photoreactive monomer, biacryloyloxy biphenyl (BAB6), cf. figure 1, and 1 wt % of the photoinitiator, benzoin methyl ether (BME). The mixture was introduced in the isotropic phase into a sandwich cell with a gap of about 2 μm. The inner surfaces of the cell substrates were covered with a conductive ITO layer and an insulating SiO_x layer. The cell was inserted in a special holder in order to apply a mechanical shear to the liquid crystal by moving one of the cell substrates with respect to the other. Applying simultaneously a unidirectional mechanical shear and a strong a.c. electric field (the dielectric anisotropy of the material has a small positive value which favours the in-plane alignment of the helix on applying an electric field across the cell), we obtained a structure with a uniformly lying helix (ULH) (figure 2). Optically, the ULH cell behaves as a uniaxial plate cut along the optic axis which for a tightly twisted cholesteric coincides with the helix axis. The electro-optic characteristics of the experimental cells were studied in a set-up described in [8], but, in addition, a light filter was used to protect the sample from any UV components from the microscope lamp which could lead to accidental premature photopolymerization of the material. Due to the very low value of the dielectric anisotropy of TI827, the ULH texture is stable (apart from helix unwinding) even at high fields ($E \approx 50\ V\ \mu m^{-1}$), and there is almost no deviation from linearity in the electro-optic response. Immediately after aligning the cholesteric/BAB6/BME mixture in the ULH texture, the electro-optic characteristics, such as the field-induced tilt angle and response times, were measured. Then we illuminated the sample with UV light (Teklite UV curing system) for 10 min, after which a polymeric network was formed in the liquid crystal volume. During this process, no significant changes in the ULH texture were observed. After illumination, we again measured the tilt angle and the response times. We found that the switching is slower (figure 3) and the tilt angle smaller (figure 4) in the polymerized system, but the changes are relatively insignificant.

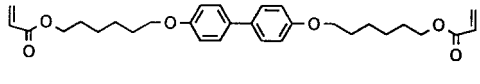

Figure 1. Structural formula of BAB6.

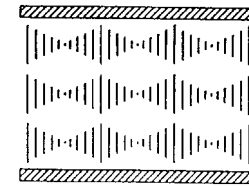

Figure 2. Uniformly lying helix (ULH) structure.

In figure 5 is shown the tilt of the optic axis and the response times as functions of field and temperature for the volume stabilized cell. The tilt is practically independent of temperature and its linearity in the field is striking. The response time is almost independent of the field strength, but decreases with increasing temperature.

The incorporated polymer structure seems to be stable with time, as four months after polymerization of the sample no degradation or relaxation of the ULH texture can be seen. On heating to the isotropic phase of TI827 ($T \approx 55$°C), the texture becomes dark between crossed

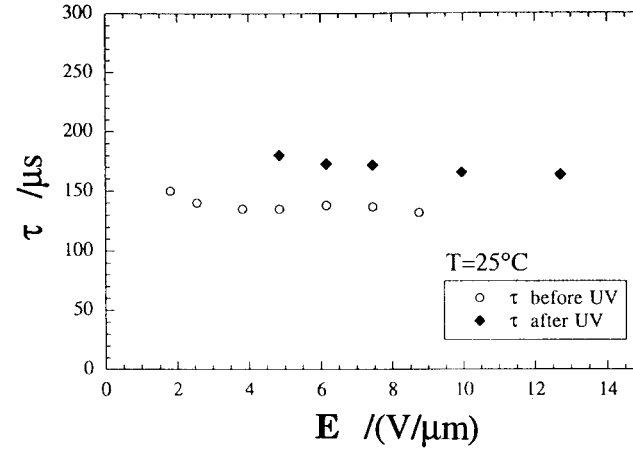

Figure 3. Response time τ before and after photopolymerization.

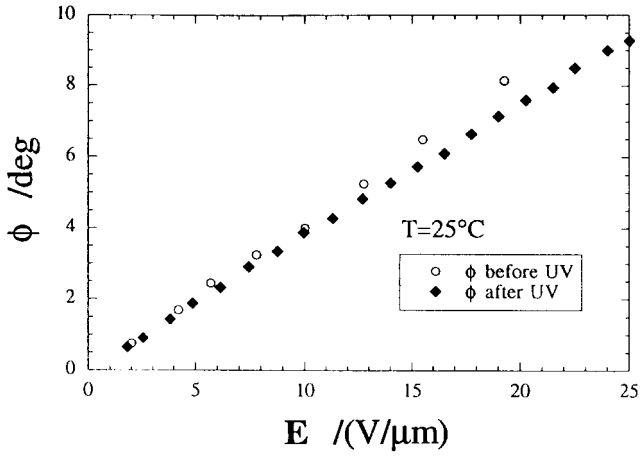

Figure 4. Field-induced tilt of the optic axis before and after photopolymerization.

polarizers, but a weak birefringence remains, indicating that the polymeric network is anisotropic. On cooling back to the cholesteric phase the ULH structure is formed again and we could see no difference in the texture compared with that before heating.

3. The cholesteric phase-shift mode

In the experiment described in § 2 we used a material with a low dielectric anisotropy ($\Delta\varepsilon$) to optimize the linearity in electro-optic response by suppressing dielectric coupling. We have also volume stabilized the ULH structure in a cholesteric with a much larger dielectric anisotropy than that of TI827, and sufficiently large for the helix to unwind at higher fields. The field strength for complete unwinding in this second material is about $20\,\mathrm{V}\ \mu\mathrm{m}^{-1}$. In the unwound state the cell is in principle completely homeotropic with the director **n** perpendicular to the glass plates, giving extinction of light between

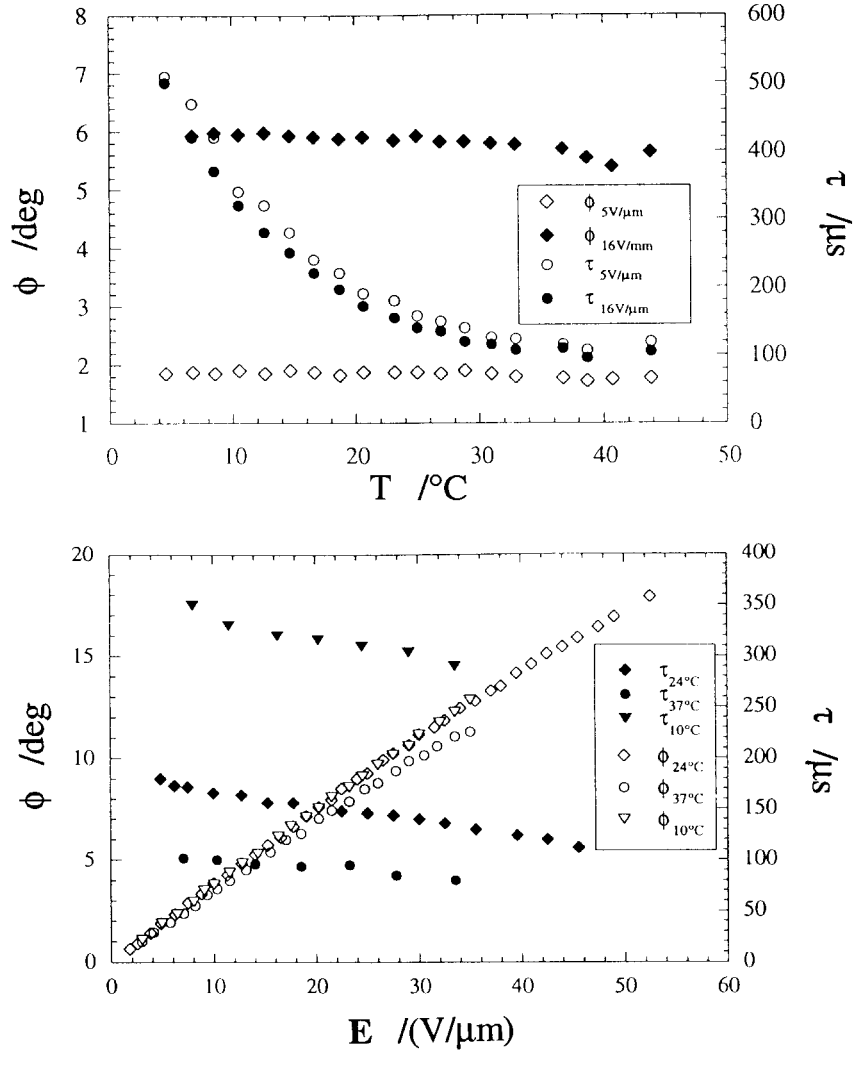

Figure 5. Electro-optic characteristics for the stabilized ULH cell.

crossed polarizers. In figure 6 is shown a photograph of the polymer stabilized ULH texture. Outside the bright electrode area with the ULH texture, we have the Grandjean texture, formed as a result of the aligning method used. As mentioned before, the ULH structure is formed by simultaneously applying an electric field and a mechanical shear. Outside the electrodes there will be no field applied and the cholesteric then adopts the Grandjean texture.

Very interesting is the fact that when the field is removed, the cholesteric twists up again and the ULH texture is recreated just as it was before unwinding. This will not happen in a non-stabilized cholesteric structure for which the removal of the field leads to a disordered focal-conic texture, cf. figure 7.

Imagine that we have a cholesteric material with a high dielectric anisotropy, but with almost zero flexoelectric response, in a polymer stabilized ULH texture. The cell behaves as a uniaxial waveplate with a certain birefringence $\Delta n_{\text{eff}} = n_{\parallel}^{\text{o.a.}} - n_{\perp}^{\text{o.a.}}$ where $n_{\parallel}^{\text{o.a.}}$ and $n_{\perp}^{\text{o.a.}}$ are related to the optic axis (which is the twist axis). If we denote the indices relative to the *director* with $n_{\parallel}$ and $n_{\perp}$, then $n_{\parallel}^{\text{o.a.}} = n_{\perp}$ in the ULH geometry and $n_{\perp}^{\text{o.a.}} \approx (n_{\perp} + n_{\parallel})/2$ as long as $E = 0$. When we turn up the field, $n_{\parallel}^{\text{o.a.}}$ is not affected, whereas $n_{\perp}^{\text{o.a.}}$ changes continuously from $(n_{\perp} + n_{\parallel})/2$ to $n_{\perp}$ at $E \geqslant E_c$, the characteristic field for complete unwinding. At this point the sample appears isotropic with respect to transmitted light. The field dependence of the birefringence of a ULH layer is studied in [7].

Consider that we make an electrode pattern on the cell glass plates providing a two dimensional array of pixels over the ULH cell area. Coherent linearly polarized laser light, with its plane of vibration perpendicular to the helix axis, will experience only the refractive index $n_{\perp}^{\text{o.a.}}$. If we now apply a voltage across some of the pixels, the light will locally experience a refractive index different from that over the pixels with no voltage applied. The difference in $n_{\perp}^{\text{o.a.}}(E)$ between different pixels will induce a phase shift ϕ due to the different optical path lengths $L = n_{\perp}^{\text{o.a.}}(E)d$ experienced and the magnitude of ϕ would depend on the voltage applied. We have in fact a field-controlled phase hologram where we can control the refractive index in a continuous manner at each pixel of the hologram (figure 8). This has obvious potential applications in Fourier optics, optical processing devices and electrically controlled kinoforms, to mention but a few examples.

The cell may be also used as a light modulator between crossed polarizers with its optic axis at a 45° angle to the polarizer axis. The effective birefringence $\Delta n_{\text{eff}} = n_{\parallel}^{\text{o.a.}} - n_{\perp}^{\text{o.a.}}$ will vary from $\Delta n_{\text{eff}} \approx (n_{\perp} - n_{\parallel})/2$ to $\Delta n_{\text{eff}} = 0$ on changing the value of the electric field. If the cell fulfils the $\lambda/2$-wave plate condition at $E = 0$, then the phase shift $\delta = 2\pi d \Delta n_{\text{eff}}/\lambda$ between the ordinary and extraordinary wave can be continuously controlled between 180° and zero and we have full modulation of transmitted light between crossed polarizers. The required fields for total unwinding of the ULH structure in our case are rather strong, about 25 V μm^{-1} in a 2 μm cell and the switching is rather slow (in the ms region) in comparison with the flexoelectro-optic effect.

4. Discussion and conclusions

We have shown that a 10 wt % concentration of photoreactive monomer forms a stabilizing polymeric network in the volume of a ULH aligned cholesteric liquid crystal on UV illumination. The problem of degradation of the ULH texture and also relaxation into the Grandjean texture is hereby solved and surface

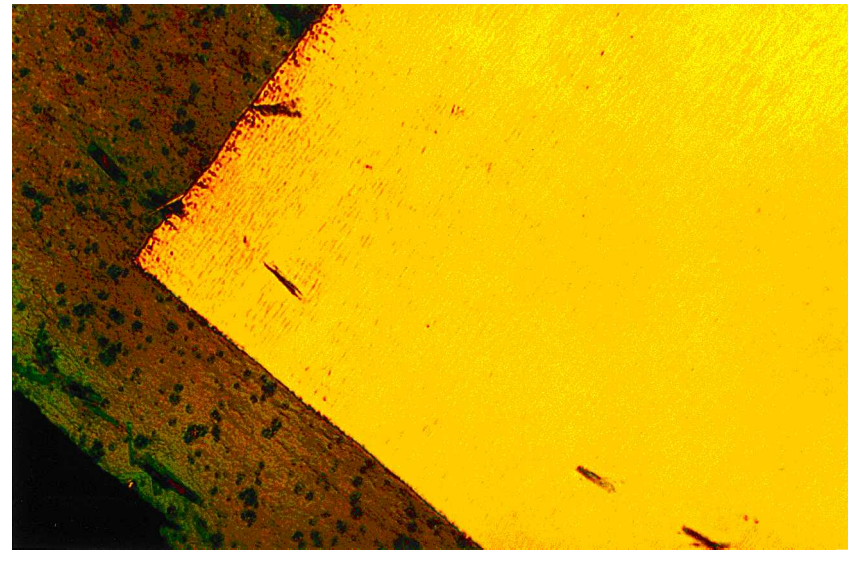

Figure 6. Photopolymerized ULH structure for the phase-shift effect.

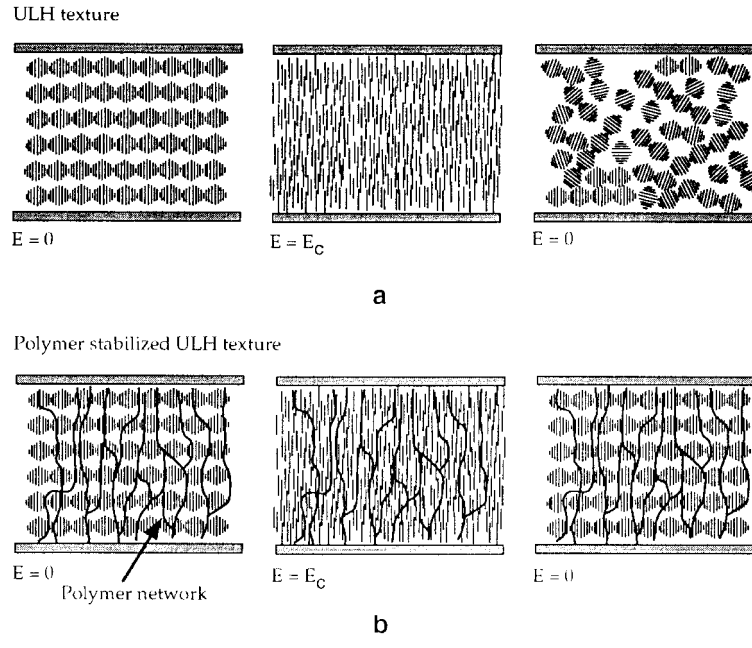

Figure 7. Field-induced untwisting of the helix in the case of (*a*) non-stabilized and (*b*) stabilized ULH texture. In (*b*) the ULH structure is recreated after removal of the electric field.

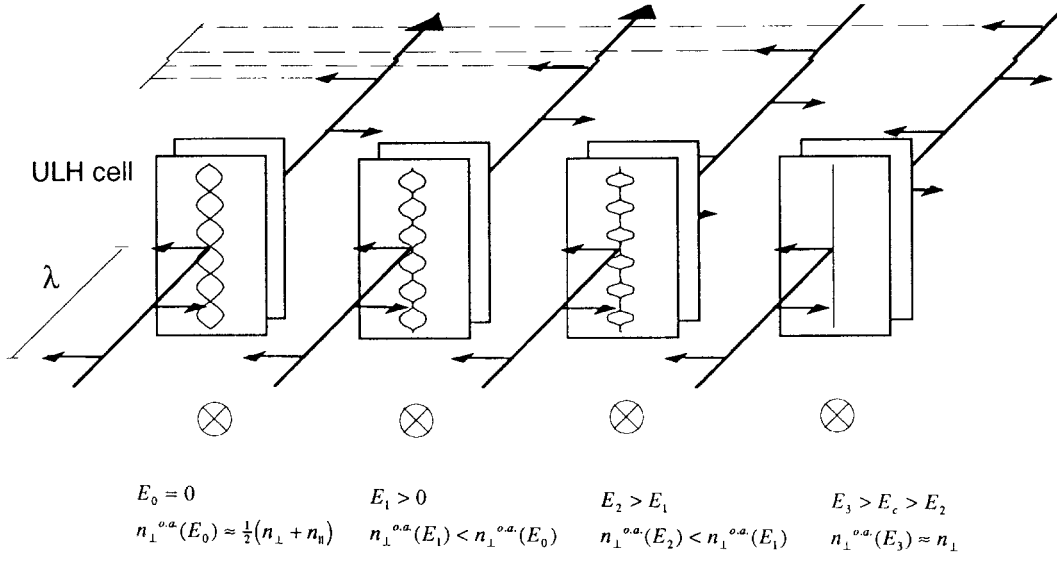

Figure 8. The phase-shift effect. Horizontally polarized light experiences an electric field-dependent effective refractive index $n_\perp^{o.a.}(E)$ depending on the degree of unwinding of the helix at each pixel.

effects, dielectric coupling and phase transitions can no longer destroy the texture.

Depending on the cholesteric used, the volume stabilized ULH structure may be utilized in two different effects: the flexoelectroptic effect and the field-induced unwinding effect. A material with a large $e = \frac{1}{2}(e_s + e_b)$ and small dielectric anisotropy $\Delta\varepsilon$, gives a linear light modulator utilizing the flexoelectro-optic effect. The formed network only slightly decreases the speed and magnitude of the response compared with the non-polymerized sample and the lowest concentration of reactive monomer needed to stabilize the ULH structure is still to be determined. If we choose a cholesteric with large $\Delta\varepsilon$ and zero e we can partly unwind the helix by means of the electric field and thereby control the effective refractive index perpendicular to the helix axis and hence also the macroscopic birefringence of the material. Even if the helix is completely unwound it goes back to the original ULH state on removal of the field. Such a device may be used as a phase modulator (phase hologram) or a light intensity modulator depending on the geometry of the experimental set-up.

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