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Alignment and switching of nematic liquid crystals embedded in porous chiral thin films

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Porous thin films with engineered microstructures have been fabricated using glancing angle deposition (GLAD). GLAD films with chiral microstructures have been previously shown to exhibit unique chiral optical response. The pores of these films were embedded with (non-chiral) nematic liquid crystals (LCs) to produce a new composite optical material wherein the GLAD film induces chiral nematic-like LC orientation. We demonstrate here reversible electro-optic switching of the LC component of these hybrid films. Unaddressed, cells of GLAD/LC hybrid films show enhanced chiral optic response compared with the unfilled GLAD film. When addressed, the chiral optic response vanishes.

1. Introduction

Critical to the optimization of optical properties in liquid crystal (LC) devices is control over the long-range orientation of the LC molecules. A variety of techniques is used to produce alignment in LC devices such as displays. One well-known technique is rubbing of the substrates [1] which causes nematic LCs near the substrate to orient parallel to the direction of rubbing. The twisted nematic liquid crystal display (LCD), as an example, has the rubbing direction of the two substrates perpendicular to each other, generating a 90° twist in the nematic orientation within the display cell.

Another technique for controlling LC orientation is the use of obliquely deposited thin films as 'tilt' alignment layers at the substrates [2, 3]. Because they influence LC orientation at the substrate surfaces only, an inherent limitation with substrate rubbing and orientation layer techniques is the inability to maintain the desired LC orientation in thicker cells, particularly with chiral or cholesteric LCs (CLC) [4], leading to loss of orientation after electro-optic addressing (that is, irreversible switching). For better control over LC alignment, a technique which induces LC alignment throughout the cell is required, rather than influencing the LC orientation near the substrate surfaces only. This has been accomplished, for instance, by phase separation in polymer-dispersed liquid crystals.

Recently, Robbie *et al.* [5] demonstrated the use of porous engineered thin films as LC alignment structures. These films, which span the volume of the cell, are fabricated by glancing angle deposition (GLAD) [6, 7], a physical vapour deposition technique which allows the fabrication of highly porous thin films with columnar microstructure controllable on the sub-micrometre scale (figures 1 and 2). GLAD uses highly oblique or glancing angle deposition (typically at angles of $\alpha > 75°$, measured relative to the substrate normal) to accentuate the atomic shadowing effects, leading to thin films with porosities tunable from 10% to 90% [8, 9]. Computer control of substrate motion during deposition [6, 10–12] allows the shape of the columnar film microstructure to be tailored as desired. GLAD has been demonstrated with a wide range of materials and several deposition processes including evaporation [6, 9], sputtering [13], and recently pulsed laser deposition [14].

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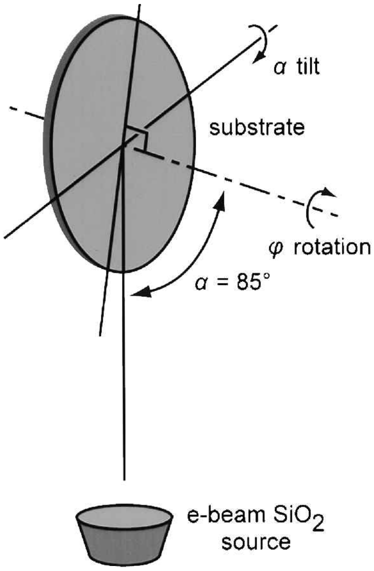

Figure 1. Schematic of glancing angle deposition (GLAD) process: stepper motors control the deposition angle (*α* tilt) and rotation of the substrate about an axis normal to its surface (*φ* rotation). A computer-based feedback control system actuates the motors in accordance with parameterized programs of *α* and *φ* as functions of accumulated film thickness.

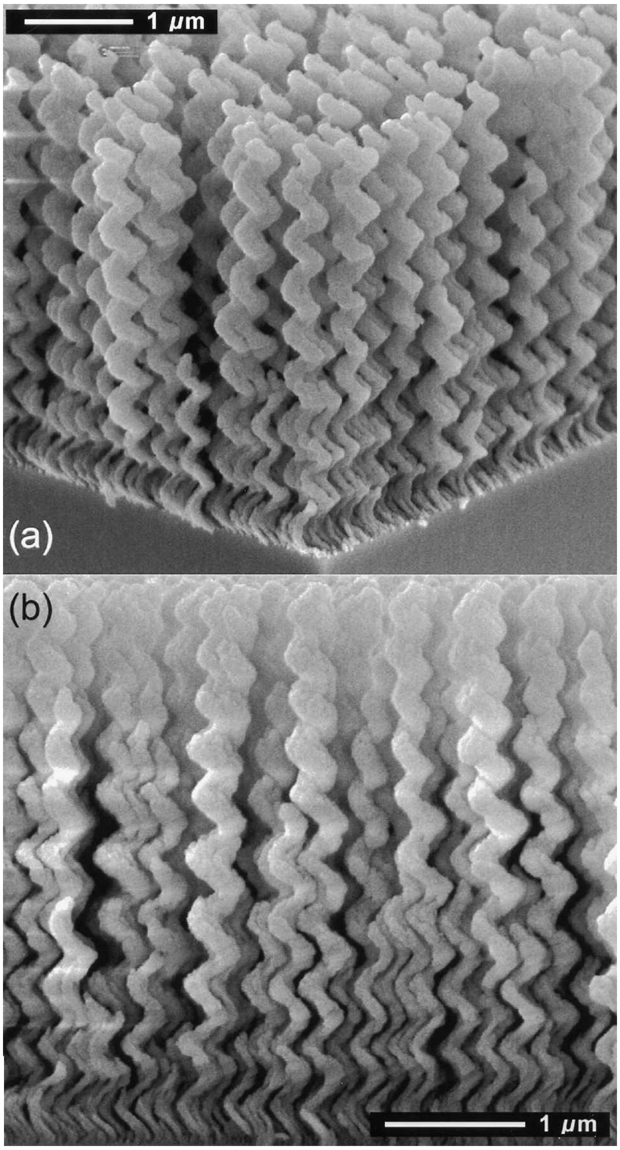

Figure 2. Examples of helical GLAD films (by SEM). The thickness of one turn of the helix is called the pitch and is related to the wavelength of peak chiral optic effects. (*a*) Left-handed helical, 8.4 turns, pitch 410 nm. (*b*) Left-handed helical, 10 turns, pitch linearly graded from 350 nm (at substrate) to 500 nm (top of film).

Of particular interest for optical applications are the GLAD films possessing 'helical' or chiral microstructures, such as those shown in figure 2. These films have been shown to exhibit optical activity and circular Bragg reflection similar to CLCs. This phenomenon was observed as early as 1959 by Young and Kowal [15] who deposited films at oblique angles ($\alpha \leqslant 70°$) onto rotating substrates and demonstrated optical activity in these films. Following theoretical work by Azzam [16] and Lakhtakia and Weiglhofer [17] which revived interest in the field, Robbie *et al.* fabricated the first porous helical thin films using GLAD [18] and subsequently reported spectroscopic measurements of optical activity [19, 20]. The unique chiral optic response of chiral GLAD films arises from the structural chirality and the index difference between the film material and the void material (air).

Robbie *et al.* [5] embedded helical GLAD films with several compounds including isotropic liquids, polymers, and reactive and non-reactive liquid crystalline materials. Circular dichroism was measured as differences between transmission of left- (LCP) and right-circularly polarized (RCP) light. Filling with isotropic liquid (e.g. water) or polymer caused all transmission difference to be lost, as these fluids were approximate index matches for the film material. More importantly, filling GLAD films with anisotropic materials such as nematic liquid crystals was found to enhance the transmission difference. Here, the helical GLAD film acted as an alignment structure, and was found to impose a chiral nematic-like ordering in the LC component.

Apart from the enhanced chiral optic response, embedding porous chiral GLAD films with liquid crystals has several other advantages. The quantity of diffuse scattering observed in unfilled films is substantial and results in low transmission. These films, especially thicker samples, appear cloudy to the naked eye and resemble frosted glass. The refractive index mismatch between the film (typically $n \sim 1.4$) and void (i.e. air, $n = 1.0$) materials and the nanoscale roughness, as observed in the SEM images of the films (figure 2), contribute to the scattering. After filling with LC, however, the transmission increases significantly, primarily due to the smaller index difference between the film and LC components.

A second advantage of embedding GLAD films with LCs is the mechanical and environmental stability offered by a sealed LC cell package. The LC cell package provides mechanical protection for the GLAD film, while the LC which fills the pores of the film provides stability against humidity and other environmental effects. The effects of humidity on semi-porous obliquely-deposited optical thin films is being studied extensively by Hodgkinson *et al.* [21].

The ability to switch the liquid crystal component is critical to the development of switchable optical devices based on GLAD thin films. Thus our current pursuits are twofold: to investigate further the effects of embedding LCs into GLAD films, and also to demonstrate electro-optic switching of the LC. In this report, we present the first demonstration of reversible electro-optic switching of the LC component in cells fabricated with chiral GLAD films embedded with nematic LCs.

2. Experimental

2.1. *GLAD thin film fabrication*

Porous, chiral GLAD films of SiO_2 ($n = 1.47$) were deposited by electron-beam evaporation onto glass substrates (figure 1), some of which were coated with indium tin oxide (ITO) transparent conductor for fabrication of the LC switching cells. The film shown in figure 2(*a*) is composed of left-handed helical columns with 8.4 turns and pitch $p = 410$ nm. The pitch is the vertical distance between adjacent coils of the helix, that is, the spatial periodicity along the helical axis. The pitch in a helical GLAD film is controlled by the ratio of deposition rate to substrate rotation rate, while the handedness is controlled by the direction of rotation. Measurements were also performed on a second film, shown in figure 2(*b*), which is left-handed with 10 turns and has a deliberately introduced gradient in the pitch from 500 nm at the substrate to 350 nm at the top of the film. Pitch gradation is one technique used to broaden the reflection band with CLCs [22].

2.2. *Cell fabrication*

Liquid crystal switching cells (figure 3) were constructed by using a second ITO-coated glass substrate to form a 'sandwich' with the GLAD film in between. UV-cure epoxy was used to seal the edges of the cell, leaving a small fill port on one edge of the structure. The two glass substrates were offset slightly to permit electrical contact to the ITO layers.

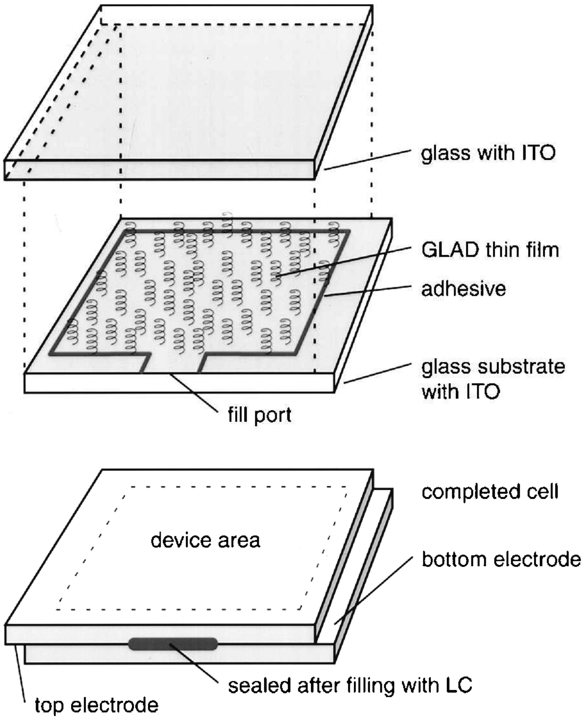

Figure 3. LC switching cell fabrication. A GLAD film is deposited on an ITO-coated glass substrate (1 inch square); a second ITO/glass substrate is used to 'sandwich' the GLAD film. UV-cure epoxy is used to seal the edges of the structure, leaving a small port for LC filling. The glass substrates are offset slightly to facilitate electrical contact to the ITO electrodes.

To fill the film with LC (nematic E7 from E. Merck, $n_o = 1.5216$, $n_e = 1.7462$ at 25°C), the cell was suspended with the fill port facing down in a vacuum oven heated to 80°C (above the isotropic clearing temperature of the LC, 60.5°C) and evacuated with a small rotary vacuum pump. The cell was then dipped into a small pool of the LC which filled the cell by capillary action. After the cell was filled, the fill port was sealed off with UV-cure epoxy.

3. Results

3.1. *Embedding of liquid crystals*

Spectroscopic circularly polarized transmission measurements were first performed on the chiral GLAD films with and without embedded LC. A Unicam 8700 series UV/Vis spectrophotometer was used to perform the measurements. The light path consisted of: light source, depolarizer, linear polarizer, quarter wave retarder oriented at ± 45° to the polarizer axis, sample, depolarizer, and detector. Left- or right-circularly polarized light was produced by the combination of the linear polarizer and the quarter wave plate (560 nm). Transmission was measured for both LCP and RCP.

Transmission spectra of LCP and RCP through the GLAD film of figure 2(*a*) is shown in figure 4(*a*), before ('film') and after ('film + LC') filling the film pores with E7. LCP has lower transmission than RCP through this sample, consistent with the LCP being preferentially scattered by the left-handed film. After filling with LC, the % transmission is higher for both RCP and LCP, consistent with reduced light scattering observed upon embedding the LC. The transmission difference between RCP and LCP, normalized to RCP transmission and expressed as %, is shown in figure 4(*b*) and shows a marked increase after filling with LC (from 5% to 84%).

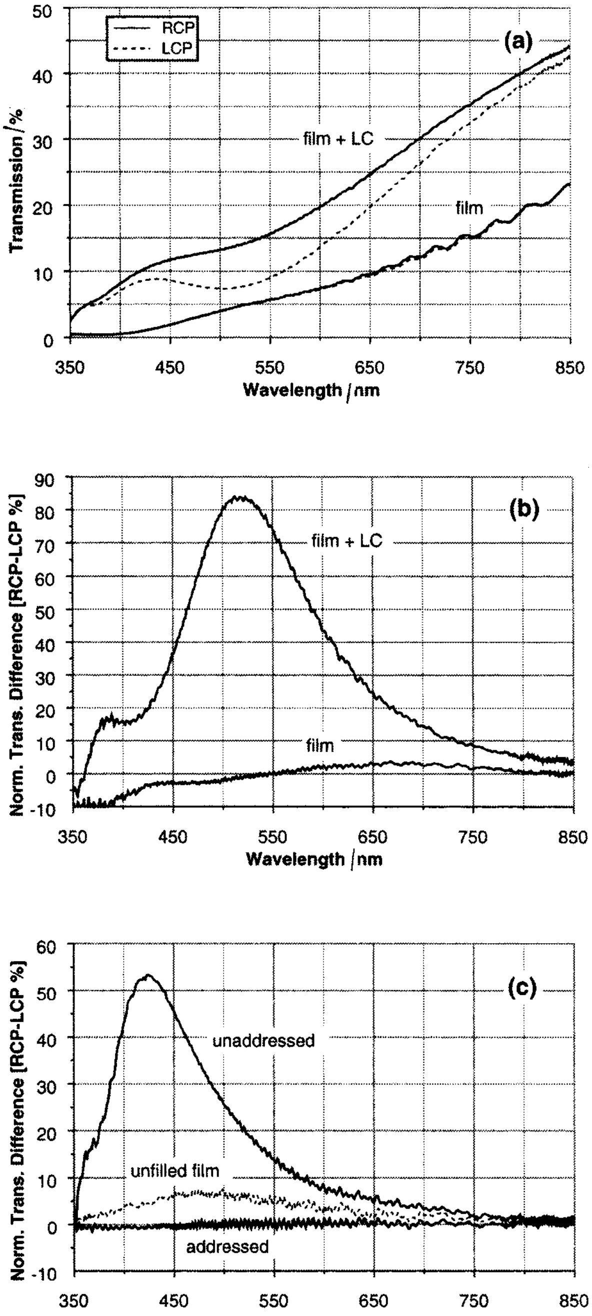

Figure 4. Circular dichroism measurements (*a*) Transmission of LCP and RCP through film of figure 2(*a*), before and after embedding the film with E7 LC. (*b*) Transmission difference RCP–LCP, normalized to RCP transmission; the addition of LC results in enhanced chiral optic response. (*c*) Electro-optic switching of LC embedded into the GLAD film shown in figure 2(*b*); when the cell is addressed, the transmission difference vanishes.

The enhanced circular dichroism suggests that a quasi-chiral nematic phase is induced in the LC by the presence of the chiral GLAD 'backbone' [5]. The LC molecules tend to align into a phase which minimizes potential energy. The effect of the chiral GLAD film is similar to that produced by the addition of chiral dopants to achiral nematic LCs.

3.2. *Electro-optic switching*

A GLAD/LC switching cell was fabricated from the film in figure 2(*b*) embedded with E7 LC. Measurements of LCP and RCP transmission were performed on this cell in addressed and unadressed modes, figure 4(*c*). In the unaddressed state (no voltage applied), enhanced circular dichroism as compared with the unfilled film is observed ('unaddressed' versus 'unfilled film'). A remarkable effect was observed when the cell was addressed by the application of a 200 V (peak-to-peak), 1 kHz signal. In the addressed state, all transmission difference between LCP and RCP vanished ('addressed'). The application of the electric field causes the LC molecules to change their alignment from the quasi-chiral nematic phase to orient parallel to the field. Transmitted light through the cell thus 'sees' the ordinary index of the LC, n_o (1.52 for E7), which is an approximate index match for the GLAD film material (SiO_2, $n = 1.47$), causing the chiral nature of the film to be effectively cancelled and resulting in the loss of circular polarization transmission difference. When the electric field was removed, the cell reverted to its unaddressed state.

4. Conclusions

Using glancing angle deposition, porous chiral thin films with tailourable microstructure may be fabricated. These films have been filled with achiral nematic liquid crystals and have been shown to induce alignment of the LC in a chiral nematic-like phase. Chiral GLAD films embedded with LCs exhibit enhanced transmission difference between left- and right-circularly polarized light as compared with an unfilled GLAD film. We have demonstrated electro-optic switching of the LC component which extinguishes the chiral optic response of the cell when it is addressed.

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