Chiral liquid crystals in photonic device applications

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Abstract

Chiral liquid crystals exhibit band-gap structures responsive to electrical and optical fields, providing wide-ranging opportunities for photonics applications. We discuss three aspects of this technology: optics of chiral nematic devices and removal of pitch jumps; optical switching of chiral nematic materials; and using novel phases in photonic devices.

1. Introduction

Chiral nematic liquid crystals are extremely wellknown for their use in thermometry devices and other applications that map out temperature or stress fields¹. Their familiar iridescence is a result of the helicoidal structure that they adopt. Typically, the helical pitch defines the wavelength of a selective reflection band (the centre of the band-gap), while the birefringence of the liquid crystal (LC), or some combination of the birefringence and tilt for a tilted phase controls the width of the reflection. The fact that chiral LCs exhibit band-gap structures that are responsive to electrical, and on occasion, optical fields provides an opportunity for wide ranging applications in photonics including lasing cavities, switchable filters and bright, bistable devices. This paper reviews our work spanning three different aspects of this technology; device structure, optically switchable materials and novel photonic band gap systems.

The first section of the paper considers the use of chiral nematic (N*) LCs in electro-optic devices. We discuss how the optical properties of the device depend on the device structure² and show how accurate modeling can completely predict the optical properties of the devices. We also consider how the shape of the reflection peak is influenced by the polarization state of the incident light, indicating an opportunity to produce switchable filters. N* materials have already been used in lasing cavities, where the lasing occurs at one edge of the reflection band, and as optical filters. In such applications pitch

jumps are extremely undesirable as they cause a sudden change in the operating wavelength. There are several approaches to remove or control pitch jumps, including careful choice of the N* material so that there is almost no pitch variation with temperature. We describe a further approach to control pitch jumps that may be of benefit in optical applications.

The second part of the paper considers optical switching of N* materials³. The use of photochromic LCs in N* systems is of considerable interest as there is potential for the reflection band to be tuned optically rather than thermally or electrically. We describe how the interplay between mechanisms that include shifts of the order parameter, diffusion of the active material outside of the irradiation area or heating of the sample contribute to the performance of these materials.

Finally, we examine the potential for using novel LC phases in photonic devices. The selective reflection of visible light from smectic phases is described. We also present details of remarkable materials that were discovered recently^{4,5}, which exhibit ferrielectric phases with both short pitches and excellent thermal stability We discuss the now realistic opportunities for using such materials in photonics applications.

2. Experimental

The experimental studies reported in this paper are carried out in devices with a thickness of $3-20 \mu m$, constructed from Indium Tin Oxide (ITO) coated glass. Rubbed alignment layers on the inner surfaces of the glass, formed from standard materials including polyimide or polyvinylalcohol, ensure good monodomain planar alignment.

Selective reflection spectra are measured using a custom-built apparatus that has been described in detail elsewhere². Briefly, the device is viewed in

reflection using a polarizing microscope fitted with a low numerical aperture objective, allowing the light to fall at approximately normal incidence on the sample. A computer-controlled monochromator resolves the reflected light, producing reflection spectra with resolution of typically ± 0.1 nm or better. The temperature of the device is controlled by a Linkam hot stage to within ± 0.1 °C. If necessary, the device can be photographed in-situ using a digitised camera attachment on the microscope.

Once the spectra have been measured, a computer program is used to fit to the data using the Berreman 4x4 matrix methodology and a Simplex minimization routine⁴. This allows optical constants to be deduced for the materials with considerable accuracy. The program also allows spectra to be calculated where the optical constants of the material/device are known.

3. Results and discussion

3.1 Chiral nematic devices

The optical properties of N* materials are normally defined by the pitch and birefringence of the system through the equations, $\lambda_0 = np$, and $\Delta \lambda = \Delta np$. Here, p, λ_0 , $\Delta \lambda$, \overline{n} and Δn are the helical pitch, peak of the selective reflection band, reflection bandwidth, average of the refractive index and birefringence of the material respectively. Consideration of these parameters allows many of the optical properties of a device to be predicted, but the response of a device must also take into account all the optical layers of the device, especially the ITO, the device thickness and the input polarization².

Figure 1 shows the measured and fitted reflection spectra for an ITO coated device with the material BL130/131⁶. The position and width of the spectrum are broadly as defined above though in order to achieve the excellent fit to the experimental data, dispersion in the refractive index and the complex refractive index of the ITO layer have been taken into account^{7,2}. The shape of the spectrum is clearly strongly dependent on the incident polarization, an effect noteworthy here as it provides an opportunity to tune the optical response of a device through mechanisms that do not depend on the material.

The fact that devices also suffer from so-called pitch jumps limits their use in some applications. The effect results from strong planar anchoring, ensuring that only a whole number of half-pitches, N, can exist within a device of thickness d, d = Np/2. Figure 2



Fig. 1. Reflection spectra from a 2.32µm device for various angles of input polarization (Angle).

illustrates the pitch jump effect, both visually with clear, discontinuous changes in colour in a device and as they manifest in the pitch-temperature graph. The phenomenon can be mitigated to some extent by material design; the BL130/131 mixtures were designed to have a very small dependence of pitch on temperature.

There has been considerable discussion in the literature about the influence of anchoring strength on pitch jumps in N* devices⁸. Clearly, pitch jumps will occur only for strong anchoring, which is the regime for most common alignment materials. There is an opportunity to take advantage of simple geometric effects to control the number of pitch jumps in a device, as follows. It can easily be shown that the size of a pitch jump is given by $\Delta p = 2d/N(N+1)$. Putting physically realistic values into this equation, we see that for a pitch of around 350nm (giving a reflection band centred at ~560nm) and a device thickness of 5 µm, there are between 14 and 15 half pitches in the device. The difference in pitch between the two stable states in such a device is 48nm. This is easily visible. However, for a thicker device, say 10 um thick, N takes values of 28 and 29 and the pitch difference is only 24nm, while at 15µm, the pitch jumps are only 16nm. There are therefore two possible approaches in designing a device. Firstly, a thicker device (>20µm) will give less obvious pitch jumps, and provided good alignment is still attainable, this is one way in which the undesirable phenomenon can be avoided. A better way, especially for optical filters, is to combine material design and device structure to ensure that, over the operating



Fig. 2. Pitch jumps in a device, showing the effects of device thickness on the spectrum and temperature dependence of the pitch.

temperature range, no pitch jumps occur. Thus, for a system that contains BL130/131 materials for which $dp/dT \approx 50 nmK^{-1}$, choosing a device that is 5 µm thick will allow a temperature change of around 50K before a pitch jump occurs. This is a useable temperature range and allows device straightforward optimal device design.

3. 2 Optical switching of N* materials.

Photochromic LCs, that change their properties as a result of irradiation with a specific frequency of light, have generated much interest in the context of all-optical devices⁹. Several physical effects can occur as a result of photochromic switching in LCs: variation in order, caused by conformational changes in the photochromic molecules; heating, resulting from absorption of radiation; and reorientation of the director which occurs as a dynamic process¹⁰. The use of photochromic N* materials in devices is particularly attractive as this provides an opportunity to produce optically switched optical devices.

In our study of photochromic N* materials, we again made use of Merck's BL130/131 family of materials since the temperature insensitive pitch dependence allows the photochromic mechanisms to be more readily understood³. We produced a photochromic N* mixture that included 10% w/w of the photosensitive nematic material made of equal weights of 1, 2, and 5 homologues of 4-n-butyl-4'-n-alkoxyazobenzene). The nematic photochromic component undergoes an isothermal phase transition on irradiation with low power (milliwatts) 488nm light¹¹. The N* mixture exhibits selective reflection at635nm at 20°C and 655nm at 95°C. The *cis-trans* isomerisation occurs rapidly on irradiation with 488nm light. The reverse reaction takes place on a longer time scale (several minutes) and can be stimulated by 633nm radiation.



Fig. 3. The optical appearance of the irradiated region (top left) following diffusion of the nonliquid crystalline component from the region. The points on the lower graphs are deduced from the labeled regions in the top right diagram (from [3])

On irradiation, the mixture initially reflecting light at ~635nm, forms concentric colour domains, figure 3. The effect is a result of the trans-cis isomerisation of the photochromic nematic material, together with diffusion of the isomerised component away from the 150 µm diameter irradiated spot. Several points can be noted. Firstly, the colour variation can be related to the concentration of the *cis*-nematic component, and is not a result of heating. This is clear because in the host N* material dp/dT > 0 (so if the central spot was heated, the surrounding material would be 'redder'). However, the coloured bands show a decreasing pitch occurring because of a fall in the concentration of the cis-nematic component. Secondly, the pitch jumps are themselves clear as distinct rings in the sample, even though the diffusion of the switched component is continuous. These are analogous to the pitch jumps shown in figure 2, but are a result of concentration changes. Finally, the reduction in order on irradiation means that the birefringence and hence reflection bandwidth will also reduce, though with a different dependence than is caused by temperature changes.

3. 3 Novel photonic materials.

This review has so far concentrated on the use of N* materials in optical device applications, though it is also well-known that the helical structures of chiral ferroelectric and antiferroelectric phases exhibit selective reflection⁴ In smectic systems, the width of the selective reflection band reduces because of the out of plane tilt of the director, figure 4.



Fig. 4. Calcuated reflection spectra for N* (left) and ferroelectric (right) systems as a function of the number of pitches in a device.

Until recently the use of the smectic intermediate (ferrielectric) phases as band gap materials had been considered only theoretically¹², primarily because of their relative lack of temperature stability (most phases occur over ranges of $< 3^{\circ}$ C) and their long pitches (usually > 1 µm). We recently reported a series of mixtures made from a single antiferroelectric liquid crystal material and a highly chiral dopant at concentrations <10% that have remarkably stable intermediate phases, tens of degrees wide⁵. Further, the pitch of the ferrielectric (3-layer repeat) phases vary from 450 nm upwards¹³, so that selective reflection of visible light is possible from the ferrielectric phases. As can be seen from figure 4, changes in the phase structure offer an opportunity to modify the shape of the reflection spectra in chiral LC systems. The fact that new, short pitch ferrielectric materials now exist offers a series of novel materials for photonic band gap applications, especially where narrow reflection bands are desired.

4. Summary

This review has considered various ways in which chiral liquid crystal materials may be used in optical devices. The use of chiral nematic materials is wellknown, and it is clear that not only can complex device optics be accurately predicted (figure 1), but that there are interesting opportunities to produce devices without pitch jumps using standard device technology. The use of photochromic N* liquid crystals is of significant interest in all-optical devices. Consideration of the possible mechanisms allows the response of the device to be tuned, depending on how the photochromic component is incorporated in the system. For a nematic photochromic material, clear changes in order and hence reflection spectra occur. Finally, the use of ferrielectric phases as photonic band gap materials has been considered, something that has only recently become practicable. It is clear that chiral nematic materials have significant potential in optical devices, way beyond their now well-known use as thermochromic materials.

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5. References

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