Chapter 5. Cholesteric liquid crystals (CLC) and their photonic properties

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1. Structure and formation of CLC

Cholesteric (or chiral nematic) liquid crystal (CLC) resembles nematic liquid crystal (NLC) in all physical properties except that the molecules tend to align themselves into a helical structure with the helical axis perpendicular to the director, as depicted in Figure 1. The helical structure leads to a selective reflection in wavelength and circular polarization. The length over which the LC directors rotate by 2π is defined as helical pitch, which indicates the extent of twist of LC molecules.

The CLC phase forms in liquid crystalline materials possessing chirality. Some molecules, such as cholesterols, form the phase naturally but more commonly, the phase is formed by adding chiral dopants to NLC. The structure of the CLC phase can be either a left-handed or right-handed helix. An illustration of a left-handed CLC helix is presented in Figure 1 (b). The unit length of one complete rotation of the director orientation is defined as the pitch (*P*). The handedness of the CLC helix is controlled by the handedness of the chiral dopant and the spectral position of the reflection is governed by Eq. (1):

$$\lambda_b = \langle n \rangle P \qquad (1)$$

where λ_b is the wavelength of Bragg reflection, $\langle n \rangle$ is the average refractive index, $(n_e - n_o)/2$, and *P* is the pitch. Typical NLC hosts have refractive indices that range from 1.4–1.8, with an average index that is predominately around 1.6. The simplest means of controlling the position of the reflection is by directly affecting the pitch. In mixtures based on chiral dopants, the pitch is equal to:

$$P = \frac{1}{[C]\times\mathrm{HTP}} \qquad (2)$$

where [C] is the concentration of the chiral dopant and HTP is the helical twisting power of the chiral dopant. The HTP of a chiral dopant is a quantifiable parameter that is indicative of the efficiency of the chiral dopant to induce a twist in an NLC host. Typical HTP values of commercially available chiral dopants range from 8–25 μm^{-1} when concentration is by weight fraction. Another optical property of CLCs is the bandwidth ($\Delta\lambda$), defined as:

$$\Delta\lambda = \Delta n P \qquad (3)$$

where Δn is the birefringence of the liquid crystal. The birefringence of the liquid crystal, equal to the difference between the extraordinary and ordinary refractive indices, can range from 0.06–0.3 in typical NLC host materialsFrom Eq. (3), it is clear that the bandwidth of CLCs increases with increasing Δn as well as with pitch (spectral position). Typical bandwidths in the VIS-NIR region are on the order of 50–100 nm.

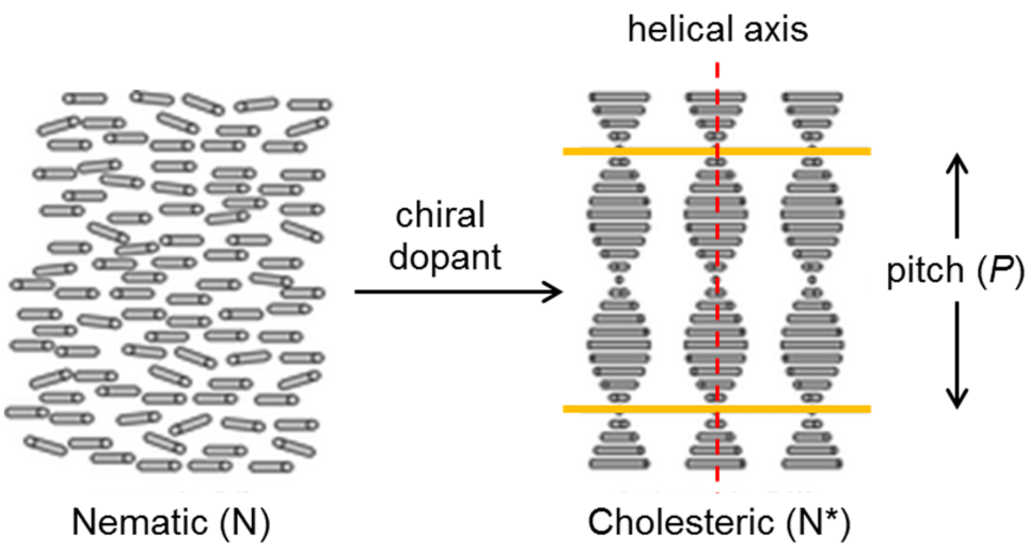

Figure 1. Illustration of the helical structure of a CLC by adding chiral dopants to NLC. As dictated by the chirality of the system, these materials form a helicoidal structure described by the pitch (P), the length for one complete rotation.

2-1 Unique optical properties of CLC

2-1 Selective light reflection

The helical structure of CLCs gives rise to the fundamental property of selective reflection of light. This phenomenon can readily be observed by the naked eye as an iridescent color when the CLC confined between glass plates presents a planar texture, for which the helical axis is perpendicular to the surfaces, and the pitch P is in the range of the wavelength of visible light. At normal incidence (i.e., for light propagating along the helical axis), the maximum of selective reflection occurs at the wavelength λ_0 which is directly related to P by Eq. (1). Circularly polarized (CP) light having the same sense as the helix twist is totally reflected, a property that is often referred to as Bragg reflection by analogy with X-ray diffraction, although only the first-order Bragg reflection is possible for normal incidence. All order reflections occur at oblique incidence or when the helical structure is distorted. The reflectance properties are very often deduced from transmittance spectra, Figure 2(a), when investigations are made with clear-cut planar oriented cholesteric samples that do not absorb or scatter light. The reflection color depends on: (i) parameters proper to the material such as the molecular chirality or the concentration of chiral dopant, the pitch, and the optical indices; (ii) external parameters such as temperature, mechanical pressure, electric or magnetic field, angle of incidence of the light ($\lambda_0 = nP\cos\theta$, where θ is the angle between the helical axis and the direction of propagation), the polarization of the incident light, or the geometry of the experimental cell.

At normal incidence, the bandwidth $\Delta\lambda$ is related to the birefringence Δn and P by Eq. (3). $\Delta\lambda$ is currently measured like the width of the bandgap at half height. $\Delta\lambda$ is limited to a few tens of nanometers in the visible spectrum because the birefringence is typically limited to 0.5. This limit is not a problem when the selectivity of the light reflection is desired for many applications such as narrow-band polarizers and filters, thermography, and, more generally, for sensors. However, the bandwidth must be dramatically increased for innovative applications like full-color or white-on-black reflective polarizer-free displays, broadband polarizers, organic optical data storage media, or smart switchable reflective windows to control solar light and heat. Recently, researchers are eager to find solutions to broaden the bandwidth around a central reflection wavelength beyond the reflectance limit

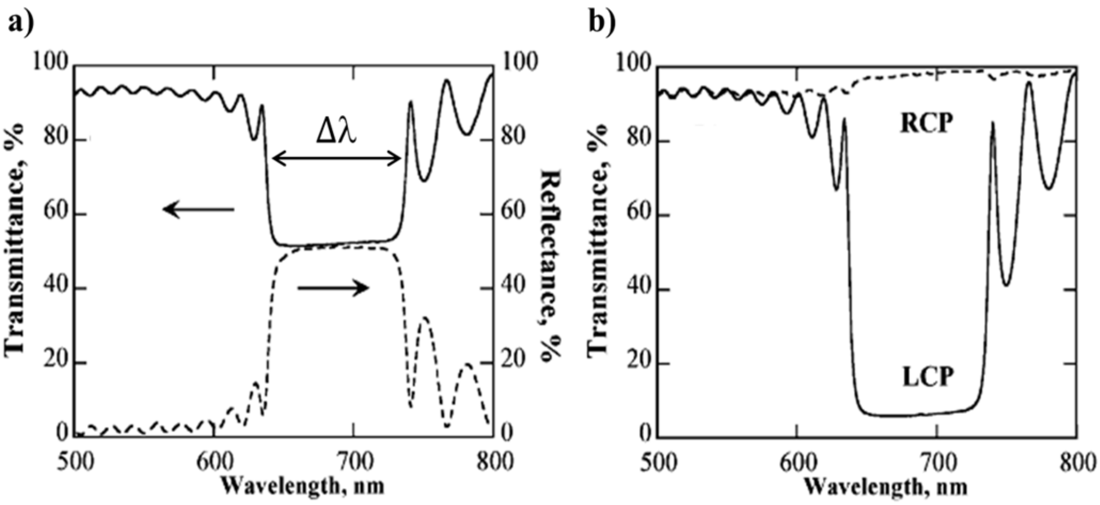

Figure 2. (a) Transmission and reflection spectra with unpolarized incident light and (b) circularly polarized transmission spectra of a 5 μm-thick CLC film between rubbed polyimide films.

2-3. Selective circular polarization of light

Consider incident light that is linearly polarized, which can be thought of as consisting of a left- and right-handed circularly polarized (CP) component. At λ_0 and normal incidence, one of these components is fully reflected by the CLC structure and the electric field pattern in the reflected wave is a helix identical in shape to the cholesteric helix, which contrasts strongly with reflection from a common mirror that undergoes a 180° phase shift upon reflection and changes handedness. The other component is simply transmitted. Right-hand circularly polarized (RHCP) and left-hand circularly polarized (LHCP) light are defined, following the usual convention, as the electric field vector rotating clockwise and counterclockwise, respectively, when viewing into the light source; that is, opposite to the direction of light propagation. The fact that the reflected light is circularly polarized with the same handedness as that of the CLC structure constitutes the polarization-selectivity rule, which is valid only at normal incidence. At oblique incidence the reflected or transmitted light is elliptically polarized. Thus, a CLC cannot reflect more than 50% of normally incident unpolarized light and reflects 0% when the incident beam is circularly polarized with a handedness opposite that of the CLC, Figure 2 (b).

This unique optical property of CLC structure has been easily observed in nature. For example, the structure on the exocuticle of the beetle, which selectively reflects left circularly polarized light and possesses a brilliant metallic appearance (Figure 3). When the left circularly polarized light is blocked by the use of a quarter wave plate and a polarizer, as shown in Figure 3, the beetle loses its characteristic bright green reflection. It is discovered that beetle cells form similar to a CLC whose free surface has cone-like structures and that has a helical arrangement of molecules. And the microstructure of the beetle surface resembles the focal conic domains of CLCs. Each of these optical mechanisms provides us with inspiration to increase the reflectivity for hyper-reflective (polarizer-free) displays, stealth technologies (also termed low-observable technologies), electro-optical glazing structures (to dynamically control electromagnetic radiation), and, more generally, polarization-independent photonic devices (since very high contrast occurs when the right- and left-handed structures reflect the same wavelength). The development for technological applications of novel CLC structures that do not obey the polarization-selectivity rule is therefore challenging.

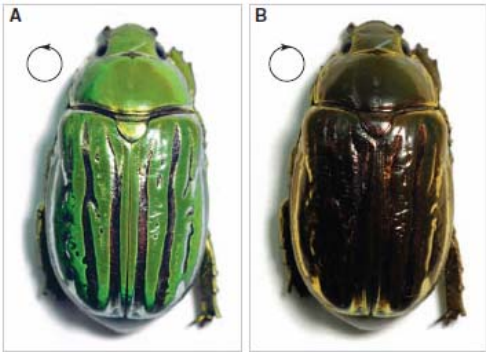

Figure 3. Photographs of the beetle *C. gloriosa*. (A) The bright green color, with silver stripes as seen in unpolarized light or with a left circular polarizer. (B) The green color is mostly lost when seen with a right circular polarizer.