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Heliconical Cholesterics: new opportunities for optofluidics?

Francesco Simoni*

Institute of Applied Sciences and Intelligent System – CNR – Pozzuoli, Italy

Abstract. In this presentation the novelty represented by the heliconical cholesteric liquid crystals (Ch-OH) for easy electric and optical control of optical properties are highlighted. After a quick summary of their electro-optical properties, an account of the recent experimental and theoretical achievements about the nonlinear optical response of Ch-OH will be given. The peculiar conical structure allows an easy control of the spectral location of the Bragg resonance making possible effects never observed previously in pure liquid crystals, making these materials attractive for development of several optical devices.

1 Introduction

It is well known that cholesteric liquid crystals (CLC) are characterized by a continuous twist in space of the director **n** that describes a helix, with **n** always orthogonal to the helix axis. Of course, such a self-assembled periodic structure gives rise to selective Bragg reflection centered at a wavelength $\lambda_B = n_{av}P$, being n_{av} the average refractive index of the material and P the pitch of the helix. Meyer[1] and De Gennes[2] in 1968 have theoretically investigated the effects of a low frequency electric field (in the following mentioned as "static field") on this structure for materials with positive dielectric anisotropy ($\Delta\epsilon = \epsilon_{\parallel} - \epsilon_{\perp} > 0$). The unwinding of the helix due to a field applied perpendicular to the helix axis was expected and early demonstrated by Khan[3]. In case of field applied along the helix direction bend and twist deformations are involved therefore the ratio of the corresponding elastic constants K_3 and K_2, $k = K_3/K_2$, strongly affects the resulting phenomenon. Usually in CLC $k > 1$ and the main effect is a rotation by $\pi/2$ of the helical axis above a threshold for the applied static field. More interesting is what happens when $k < 1$: a conical deformation takes place over a threshold field, in which the director twists and bends around the heliconical axis. The main difference with respect to the conventional cholesteric state is that the director is not perpendicular to the heliconical axis but makes with it an angle $0 < \theta < 30$. The main consequence is the continuous change of the pitch (and of the Bragg wavelength λ_B) induced by an electric field applied parallel to the heliconical axis. A few years ago, materials with $k < 1$ have been synthesized thus allowing the realization of the conical deformation of cholesterics defined as "oblique heliconical cholesterics" (Ch-OH), in this way the possibility of tuning the Bragg resonance over the whole visible spectrum has been demonstrated by the group of Lavrentovich[4-6] by varying the applied static field.

2 Light-induced reorientation and nonlinear optical properties

After the discovery of the Giant Optical Nonlinearity (GON) in nematic liquid crystals (NLC) the possibility of using light-induced reorientation of the molecular director in CLC to get all-optical tuning of the Bragg resonance was investigated. However, it was demonstrated that GON cannot be achieved in CLC[7,8] therefore no significant light-induced modulation of the helical structure has never been observed in pure cholesterics.
On the contrary Ch-OH provide a new opportunity to observe this effect due to the bend deformation of the director that leads to the conical structure. In fact, this deformation allows an efficient coupling between the optical field and the director **n**. In this case we get an optical torque competing with the one due to the static field since it pushes the director towards a direction perpendicular to the helix axis while the static field torque pushes the director towards the direction of the helix axis. The equilibrium condition is given by a conical structure where the effective static field that stabilizes the structure is intensity dependent and is lower than the applied one, thus corresponding to a different pitch of the helix[9]:

$$E_{eff}(I) = \sqrt{E^2 - CI}$$

where:

$$C = \frac{\Delta\epsilon_{OPT}}{\Delta\epsilon\ \epsilon_0 c n_{av}}$$

being $\Delta\epsilon_{OPT}$ the dielectric anisotropy at optical frequencies and c the speed of light in vacuum. As consequence, we get light-induced tuning of the Bragg resonance because the pitch of the structure depends on the intensity I according to the expression:

* e-mail: f.simoni@photomat.it

$$P(I) = k\frac{E_{NC}P_0}{E_{eff}(I)}$$

being E_{NC} the threshold field over which the structure unwinds to a uniaxial nematic and P_0 the pitch of the corresponding planar cholesteric.

This effect has been experimentally demonstrated using a pump-probe technique. The pump beam (a CW frequency doubled Nd:YAG laser at λ = 532 nm), was focused on the Ch-OH sample 20 mm thick and probed by a white light (SLED) source, focused by the same optics, while a static field was applied to stabilize the conical structure. The inverted microscope set up allowed monitoring both the reflection spectra and the sample appearance in reflection mode. At a fixed value of the static electric field a light-induced red-shift of the Bragg wavelength higher than 100 *nm* has been measured[10]. Additional proof of the optical reorientation effect has been provided by measuring the light-induced change of the polarization state of the transmitted light that corresponds to the behavior observed without pump beam when varying the applied static field. For wavelength close to the Bragg resonance polarization changes from linear to elliptical to circular and back were detected by increasing the pump beam intensity, thus obtaining an all-optical control of the probe light polarization at the exit of the Ch-OH sample.

Furthermore, the nonlinear effects on the light propagation due to optical reorientation have been investigated[9]. We have found that for wavelengths close to λ_B the induced red shift of the Bragg resonance increases or decreases the reflectivity depending on the light intensity and polarization leading to a stop band in the light transmittivity for a definite range of intensity dependent on the applied static field.

Recently a more accurate calculation of the nonlinear propagation based on Ambartsumian's layer addition modified method[11] confirmed such behavior for moderate intensities while showing the onset of optical instabilities leading to self-oscillations for increasing value of the light intensity.

These peculiar optical properties have been shown to be attractive for developing tuneable optical devices such as optical notch and bandpass filters[12] and envisage the possibility of being exploited in advanced sensing devices in optofluidic configurations[13] or coupled with piezoelectric photo-mobile supports[14].

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