Optical control of light polarization in heliconical cholesteric liquid crystals

G. Nava[1], R. Barboza[2], F. Simoni[2,3], O. Iadlovska[4], O.D. Lavrentovich[4] and L. Lucchetti[2]

[1] *Medical Biotechnology and Translational Medicine Department, Univeristy of Milano, 20054 Segrate, Italy*
[2] *Dipartimento SIMAU, Università Politecnica delle Marche, Via Brecce Bianche, 60131 Ancona*
[3] *Institute of Applied Sciences and Intelligent Systems – CNR, Via Campi Flegrei, 34 - 80078 Pozzuoli, Italy*
[4] *Advanced Materials and Liquid Crystal Institute and Department of Physics, Kent State University, Kent, Ohio 44242, USA.*

**Corresponding author: l.lucchetti@univpm.it*

Received XX Month XXXX; revised XX Month, XXXX; accepted XX Month XXXX; posted XX Month XXXX (Doc. ID XXXXX); published XX Month XXXX

We show here that light polarization of a beam propagating through a heliconical cholesteric cell can be controlled by tuning the Bragg resonance of the structure. We demonstrate that this control is achieved by varying either the low frequency electric field or the intensity of a pump beam impinging on the sample. The study confirms the recently reported phenomenon of optical tuning of the heliconical cholesterics and opens the door for the development of simple and efficient polarization modulators controlled electrically or optically.

The demonstration of the existence of the heliconical configuration of a cholesteric liquid crystal (Ch_{OH}) [1] provided new perspectives for applications of this mesophase in optical devices due to the easy control of the Bragg resonance in a broad spectral range [1-3]. Recently we have demonstrated how such a structure can be efficiently exploited for optical filtering either in notch or bandpass configuration [4]In the latter we have exploited the strong optical rotatory power of Ch_{OH}, which is similar to that of a conventional right-angle cholesteric liquid crystals (CLC) [4].

Here we report how the strong optical rotatory power of Ch_{OH} could be used to control a polarization of propagating light by applying either a static electric field or an optical field. The optical control is based on the tuning of the heliconical pitch [5] by a pump beam and is thus conceptually similar to the control by a low-frequency electric field [1-4], The effect enables an all-optical control of light polarization.

A conventional CLC helicoidal structure with the local director perpendicular to the axis of twist leads to a Bragg reflection for light wavelengths in a narrow range around $\lambda_B = n_{av}P$, where n_{av} is the average refractive index of the material and P is the pitch of the structure. Light propagates in CLC as a superposition of two waves circularly polarized in opposite directions (clockwise and counterclockwise). The wave polarized with the same handedness as the helix suffers a strong Bragg reflection if its wavelength λ is close to λ_B, while the other one is almost unaffected [6-9]. If the incident light is linearly polarized, a strong change of the polarization state is expected in the transmitted beam because of the disbalance between the two circularly polarized components; a strong optical rotation is observed for the transmitted light. The rotatory power ρ for light travelling along the helix can be written as [8]:

$$\rho = \frac{2\pi\alpha^2}{8P\left(\frac{\lambda}{\lambda_B}\right)^2\left[1-\left(\frac{\lambda}{\lambda_B}\right)^2\right]} \qquad (1)$$

where

$\alpha = \frac{\epsilon_\parallel - \epsilon_\perp}{2n_{av}}$, $\epsilon_\parallel$ is the dielectric permittivity in the direction parallel to the molecular director and $\epsilon_\perp$ is the one in the direction perpendicular to it;$n_{av} = \sqrt{(\epsilon_\parallel + \epsilon_\perp)/2}$.

In Ch_{OH} a low frequency electric field induces a stable heliconical configuration of the director while preserving a single-harmonic structure. As already pointed out [10] all the results concerning light propagation along the helix of a CLC can be applied to a Ch_{OH} as well, making a few simple changes in the mathematical description. While in CLC the molecular director is orthogonal to the helix axis ($\theta = \pi/2$), in Ch_{OH} it makes an angle dependent on the applied electric field, usually in the range $0 < \theta < \pi/6$. This tilt leads to an effective value of the dielectric permittivity that depends on the angle:

$$\epsilon_{eff} = \frac{\epsilon_\parallel \epsilon_\perp}{\epsilon_\parallel - (\epsilon_\parallel - \epsilon_\perp) sin^2\theta} \qquad (2)$$

With $\epsilon_\parallel$ replaced byϵ_{eff} Eq.(1) becomes valid also for Ch_{OH}. This means that tuning of the Bragg wavelength λ_B by an electric field, as reported in [1-4], would produce a strong variation of the polarization state of a light beam of a wavelength λ close to λ_B

and. In addition, we have recently demonstrated that an optical field can exert a torque on the molecular director of Ch_{OH}, pushing it away from the helix axis and increasing the pitch, thus inducing a red-shift of the Bragg resonance [5]. Therefore, we may expect that such an optical field is able to modify the polarization state of a weak beam travelling through a Ch_{OH} cell when the intensity dependent $\lambda_B(I)$ approaches λ.

The Ch_{OH} material has been obtained by mixing two dimeric liquid-crystal (LC) 1",7"-bis(4-cyanobiphenyl-4'-yl) heptane (CB7CB) and 1",11"-bis(4-cyanobiphenyl-4'-yl)undecane (CB11CB), rodlike mesogen pentylcyanobiphenyl (5CB) (Merck) and left-handed chiral dopant ZLI-811 (Merck), in weight proportion 5CB:CB7CB:CB11CB:ZLI811 = 49.3:30,2:15,5:5 [10]. The individual flexible dimers CB7CB and CB11CB were obtained from various sources: Department of Chemistry, School of Natural and Computing Sciences, University of Aberdeen, Scotland, United Kingdom (E. Cruikshank, G.J. Strachan, J.M.D. Storey, and C.T. Imrie), Department of Chemistry, University of Hull, United Kingdom (C. Welch and G. Mehl), Organic Synthesis Facility at Advanced Materials and Liquid Crystal Institute, Kent State University, Ohio (H. K. Bisoyi, H. Wang, and Q. Li), Air Force Research Laboratory, Wright Patterson Air Force Base, Ohio (M. Rumi and T. J. White). The components from different sources show similar properties. The Ch_{OH} structure with a left-handed helix exists in the range (21– 57)°C. The mixture was sandwiched between two conductive glasses treated to promote planar alignment. The cell thickness is 21.5 µm, as fixed by Mylar spacers and measured by an interferometric technique. The heliconical cholesteric structure is stabilized by a low frequency electric field (square wave, 5 KHz; we refer to it as a "static" field in what follows) up to the maximum value 4.5 $V/\mu m$, above which the helix is unwound. For the studied material, the pitch P_0 of the cholesteric state is 590 nm.

Measurements of the state of polarization of the light travelling through the cell were performed at the controlled temperature T = 25°C, using the experimental set-up sketched in Fig. 1. A low power monochromatic beam from a laser diode ($\lambda = 405\ nm$) was used as a probe and impinged at normal incidence on the LC cell. In the first experiment, aimed at demonstrating the electric field-induced control of light polarization, the static electric field was varied in the range (2.4 – 3.0) V/µm. In the second, the optical control of the probe beam polarization was studied by fixing the static field at $E = 5\ V/\mu m$ correspondent to unwound helix. Under these conditions, a pump beam ($\lambda = 532\ nm$) linearly polarized orthogonal to the helix axis, collinear to the probe and of intensity in the range $(10 \div 16)x10^3 W/cm^2$, was used to reestablish the Ch_{OH} structure and tune the pitch, as described in [5]. Note that the wavelength of the probe beam corresponds to a short pitch that, according to [5] is easily restored by the pump beam starting from the unwound configuration.

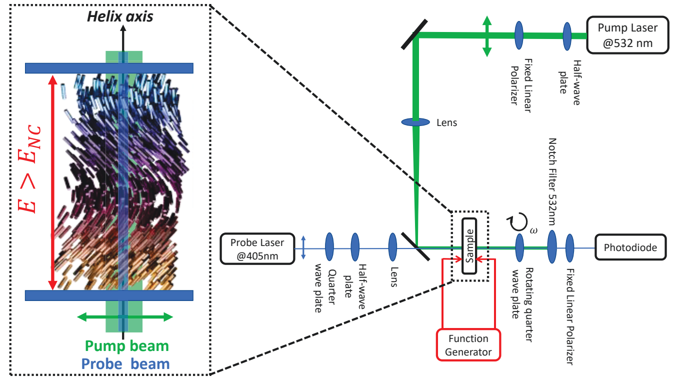

Fig.1. Sketch of the experimental apparatus for polarimetry measurements (see text), and blow out of the LC cell with pump and probe beams and static electric field.

The polarization of the probe beam was varied among linear horizontal, linear vertical, circular righthanded and circular lefthanded and was controlled by three optical elements: a Glan-laser polarizer (to stabilize the polarization state of the laser), a quarter wave plate and a half wave plate. An additional lens focuses the laser beam on the sample to a waist of 300 µm. The polarization of the light transmitted by the Ch_{OH} cell was analyzed by a rotating quarter waveplate, a fixed Glan-laser polarizer and a photodiode. The rotation of quarter-waveplate was precisely controlled by a URS100CC rotation stage and an ESP300 controller (both from Newport) with an accuracy of 0.1 deg. A custom Labview program was used to record the transmitted probe light intensity I_P versus the angle α (α=0 identifies the direction parallel to the optical axis of the plate). Cell temperature was controlled by a CalTek sample holder with an accuracy of 0.1 °C. To avoid artifacts due to mechanical misalignment, a preliminary baseline measurement with the sample in homeotropic configuration – obtained by fixing the field $E = 5\ V/\mu m$ and no pump illumination - without the output linear polarizer, was performed. In these conditions, the polarization of the light travelling through the cell stays unchanged and any variation of $I_P(\alpha)$ is to be ascribed to stirring of the beam or to a small dichroism of the rotating quarter waveplate. Each measurement was then divided by the baseline signal. A similar preliminary check was performed in the presence of the output polarizer to verify the quality of the polarization state of the probe light impinging on the cell.
The transmission curves were fitted by the function [11]:

$$I_P(\alpha) = \frac{1}{2}[A - Bsin(2\alpha) + Ccos(4\alpha) + Dsin(4\alpha)] \quad \mathbf{(3)}$$

where the fitting parameters $A, B, C,$ and D are related to the Stokes parameters describing the output polarization state [11]:

$$S_0 = A - \frac{C}{tan^2\left(\frac{\delta}{2}\right)} \quad \mathbf{(4a)}$$

$$S_1 = \frac{C}{sin^2\left(\frac{\delta}{2}\right)} \quad \mathbf{(4b)}$$

$$S_2 = \frac{D}{sin^2\left(\frac{\delta}{2}\right)} \quad \mathbf{(4c)}$$

$$S_3 = \frac{B}{sin(\delta)} \quad \mathbf{(4d)}$$

Here δ is the phase delay introduced by the quarter waveplate at $\lambda = 405\,nm$, that has been measured to be $118.2\,deg$. The retrieved Stokes coefficients allow one to obtain the polarimetric curve for each case under investigation.

The first set of measurements has been performed without the pump beam in order to characterize the effect of the applied static field on the polarization of the probe beam. Given the values of E_{NC} and P_0, the electric field to be applied to get the Bragg peak at the probe beam wavelength $\lambda_P = 405\,nm$ is obtained as:

$$E_{405} = \frac{P_0 E_{NC}}{n_{av}\lambda_P} = 2.72\,V/\mu m \quad \mathbf{(5)}$$

We thus chose to vary the static field in the range $2.4\,V/\mu m < E < 3.0\,V/\mu m$.

Figures 2*a-d* show the experimentally measured transmitted probe intensity vs the rotation angle α of the quarter wave plate for different applied static fields in the mentioned range. The data are shown for different polarizations of the incoming probe beam: linear vertical (*a*), linear horizontal (*b*), circular righthanded (*c*), circular lefthanded (*d).* Dashed lines are best fits with eq.(3).

Data related to the pump probe configuration are shown in Fig. *2e-h* for the different states of polarization of the probe beam. It is evident that the data look pretty much the same in the two cases for each probe polarization, pointing out that the reorientation induced by the static electric field is similar to the one induced by the light field, but progresses in the opposite direction. In both cases we observe the expected changes in the probe intensity by rotating the quarter wave plates with a periodicity of $\pi/2$ due to variations in the light polarization which is affected in similar ways by varying either the static field or the impinging pump intensity. The only cases where no effect is detected are the ones corresponding to the right-handed circular polarization of the probe beam (Figure 2*d* and Figure 2*h*) that does not interact with the lefthanded helix of the sample.

The data in Figure 2 and Eqs.(3,4) are used to find the Stokes parameters for each angle α which determine the polarization state of the probe beam at the exit of the sample. These parameters allow one to represent the polarization ellipses for each analyzed case. Results are reported in Fig. 3 for both electrical (a-d) and optical tuning (e-h) of the Ch_{OH} pitch.

Notice that Figures 3*e-h* is very similar to Figures 3*a-d*. In the case of electrical tuning of the pitch, the maximum effect on the probe polarization is observed at E = 2.75 V/μm, in fair agreement with Eq. (5), while there is practically no effect on the righthanded circular polarized probe beam.

At fixed static field, i.e. in the case of optical tuning of the pitch, we observe the same behavior by varying the intensity in a neighborhood of the value $I_R = 12.7 \times 10^3\,W/cm^2$, that we therefore assume to be the intensity corresponding to the resonant wavelength, i.e. λ_B (I_R) = 405 nm. This quite low light intensity is easily accounted for by considering that the applied static field is just above the critical field E_{NC} for helix unwinding, thus a weak optical torque is sufficient to restore the helical structure with a short pitch.

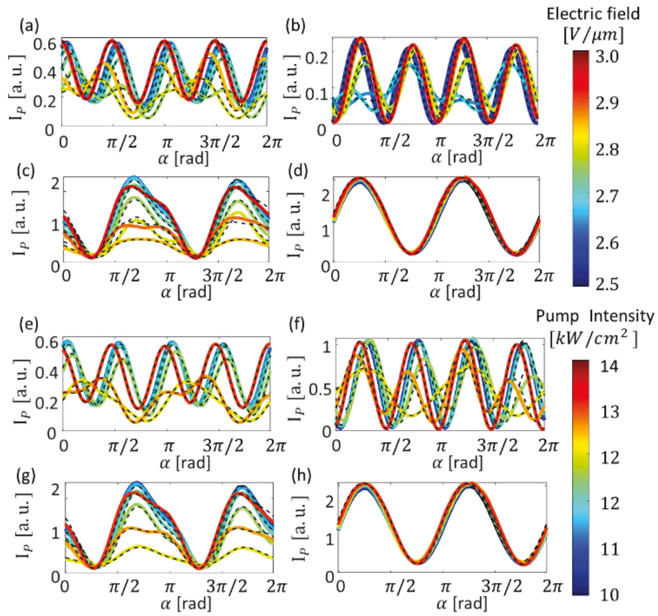

Fig.2. (a) Polarimetric measurements of the probe beam at the exit of the Ch_{OH} sample. The upper figures (*a-d*) report the transmitted signal vs the rotation angle of the quarter wave plate for different values of the applied static field (indicated by colors on the right side) with no pump beam. They correspond to different polarizations of the incoming probe beam: linear vertical (*a*), linear horizontal (*b*), circular lefthanded (*c*), circular righthanded (*d*). The lower curves (*e-h*) show the same experimental quantities obtained with a fixed value of the static field $E = 5\,V/\mu m$ while varying the pump intensity according to the color scale shown on the right side.

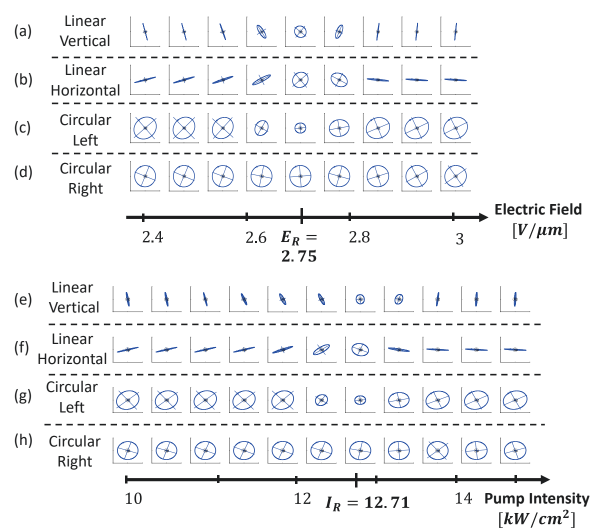

Fig.3. Plots of the polarization state of the probe beam at the exit of the Ch_{OH} sample. The upper figures (*a-d*) concern the change of the polarization state for increasing values of the applied static field with no pump beam. They correspond to different polarization of the incoming probe beam as indicated. The lower figures (*e-h*) show the changes obtained with a fixed value of the static field $E = 5\,V/\mu m$ while varying the pump intensity.

The relevance of the reported observations is manyfold. First of all, the study provides an additional proof of the recently reported optical tuning of the Bragg resonance in heliconical cholesteric liquid crystals [5]. We observe that starting from the electrically-unwound state, the heliconical structure is restored by an optical beam of moderate intensity, which imposes an optical torque on the director, overcoming the aligning action ofthe static field, as described in [5].

Secondly, we have demonstrated that the electrically-addressed Ch_{OH} film controls the light polarization by simply tuning the Bragg reflection wavelength in the vicinity of the wavelength of the propagating light beam. Such a tuning is hardly possible in a conventional right-angle CLC, since the electric field destroys the single-hadmonic periodicity of CLC.

Importantly, a similar efficient and easy to implement control of light polarization can be achieved by using an optical pump beam of moderate intensity at a fixed value of the static field. The effect enables optically controlled phase retarders of a very simple architecture. This has two important consequences: i) thanks to the efficient optical tuning of the helix pitch, the polarization state of light with any wavelength from UV to near IR could be easily controlled using the same sample and ii) the resonance wavelength can be tuned gradually by slowly varying the intensity of the pump beam thus imposing a fine control on the polarization state of monochromatic light. The amount of optical rotation of the axis of a linearly polarized beam can be tailored by choosing the appropriate value of the pump intensity (closer or farther to I_R). In the same way the transmittivity and reflectivity of a circularly polarized light can be finely controlled.

The described tuning capabilities are impossible with conventional CLCs since the electromagnetic shift of a Bragg resonance in CLCs destroys the single-harmonic structure, rendering Eq.(1) inaccurate. In principle, a similar tunning could be expected in a chiral smectic C, which shows an oblique helicoidal structure similar to that of Ch_{OH}. However, the periodic molecular-scale density modulation of smectic C (which is absent in Ch_{OH}) makes any changes of the pitch and conical tilt angle difficult since it requires to change the thickness of molecular layers and introduction of multiple defects such as dislocations with a molecular-scale Burgers vectors. In the density-uniform Ch_{OH}, the pitch change could produce only dislocations of a large Burgers vector equal P; these dislocations could be avoided or expelled if the cell is sufficiently thick, or if it is treated for a weak planar (1) or perpendicular director anchoring (2). The effects of cell thickness and surface anchoring are currently under investigation.

In conclusion, we showed that the polarization state of light travelling through a Ch_{OH} cell can be efficiently controlled by both a low frequency electric field and light. Specifically, the polarizization can remain linear but rotated by a pre-defined angle or become circular depending on the value of the electric or optical field. The mechanism of the tuning is rooted in the field-dependent pitch of the ChOH structure. An important advantage of the Ch_{OH} structure is that the electric and optical tuning tuning preserves the single-mode periodicity of the structure and thus the efficiency of polarization modification. The study opens the door for the development of phase retarders tuned either electrically or optically.

Disclosures. The author declares that there are no conflicts of interest related to this article.

Acknowledgements. ODL acknowledges the support of the US National Science Foundation grant ECCS-1906104.

References

1. J. Xiang, S. V. Shiyanovskii, C. Imrie and O. D. Lavrentovich, Phys. Rev. Lett., **112**, 217801 (2014).

2. J. Xiang, Y. Li, Q. Li, D. A. Paterson, J. M. D. Storey, C. T. Imrie, O. D. Lavrentovich, Adv. Mat., **27**, 3014 (2015).

3. M. Mrukiewicz, O.S. Iadlovska, G. Babakhanova, S. Siemianowski, S.V. Shiyanovskii and O.Lavrentovich, Liquid Crystals **46**, 1544 (2019).

4. G. Nava , F. Ciciulla , F. Simoni , O. Iadlovska , O. D. Lavrentovich, L. Lucchetti, Liquid Crystals DOI: 10.1080/02678292.2021.1884911 (2021).

5. G. Nava, F. Ciciulla, O.S. Iadlovska, O.D. Lavrentovich, F. Simoni, L. Lucchetti, Phys. Rev. Res. **1**, 033215-5 (2019).

6. P.G. de Gennes, *Physics of Liquid Crystals*, Clarendon Press, Oxford, 1975

7. V.A. Belyakov, V.E. Dmitrienko, *Optics of Chiral Liquid Crystals*, Harwood Academic, London,1989.

8. L.Blinov, *Structure and Properties of Liquid Crystals*; Springer, Dordrecht, 2011.

9. P. Yeh, C. Gu, *Optics of Liquid Crystal Displays*, Wiley, Hoboken, 2010.

10. F. Simoni, Optics Letters **45**, 6510 (2020).

11. R. Chipman, W. Lam, and G. Young, *Polarized Light and Optical Systems*, Optical Sciences and Applications of Light, CRC Press, Boca Raton, 2018.