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Authors:	G. Nava, R. Barboza, F. Simoni, O. Iadlovska, O. D. Lavrentovich, and L. Lucchetti
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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, University 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*

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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}) in 2014 [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 crystal (CLC) [4].

Here we report how the strong optical rotatory power of Ch_{OH} could be used to control the polarization of propagating light by applying either a low frequency electric field or an optical field. The optical control is based on the tuning of the heliconical pitch by a pump beam [5] and is thus conceptually similar to the control by a low-frequency electric field [1-4]. The effect enables an efficient 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 and a strong optical rotation is observed for the transmitted light. The rotatory power ρ for linearly polarized light travelling along the helix can be written as [8]:

$$\boldsymbol{\rho = \frac{\pi P(n_{\parallel}^2 - n_{\perp}^2)^2}{8\lambda^2(n_{\parallel}^2 + n_{\perp}^2)\left[1 - \left(\frac{\lambda}{\lambda_B}\right)^2\right]}} \qquad (1)$$

where $n_{\parallel}$ is the refractive index in the direction parallel to the molecular director and $n_{\perp}$ is the one in the direction perpendicular to it.

In the recently observed 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. Indeed, 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 refractive index that depends on the angle:

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

The dependence of the refractive index on θ and thus on the electric field that stabilizes the structure, allows an efficient and wide electrical tuning of the pitch, as reported in [1-4], never observed with conventional CLC. With $n_{\parallel}^2$ replaced by n_{eff}^2 Eq.(1) becomes valid also for $Ch_{OH.}$ This means that tuning of the Bragg

wavelength λ_B by an electric field, would produce a strong variation of the polarization state of a light beam of wavelength λ close to λ_B. In addition, we have recently demonstrated that, thanks to their oblique configuration, an optical field can exert a torque on the molecular director of $Ch_{OH,}$ pushing it away from the heliconical 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: University of Aberdeen (E. Cruikshank, G.J. Strachan, J.M.D. Storey, and C.T. Imrie), University of Hull (C. Welch and G. Mehl), Kent State University (H. K. Bisoyi, H. Wang, and Q. Li), Air Force Research Laboratory (M. Rumi and T. J. White). CB7CB obtained from different sources shows the same temperatures (within 1 - 1.5 °C) of phase transitions between the isotropic, nematic, and twist-bend nematic phases. The same is true for CB11CB. 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) up to the maximum value $E_{NC} = \frac{2\pi}{P_0}\sqrt{\frac{K_2}{\varepsilon_0 \Delta\varepsilon K_3}} = 4.5\ V/\mu m$, above which the helix is unwound. For the studied material, the pitch P_0 of the cholesteric state is 350 nm.

Measurements of the state of polarization of the light travelling through the cell were performed at the controlled temperature T = 25°C, at which the liquid crystal birefringence is 0.2 [11], 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 low frequency 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 electric field at $E = 5\ V/\mu m$ correspondent to unwound helix. Under these conditions, a pump beam ($\lambda = 532\ nm$) with Gaussian profile, linearly polarized orthogonal to the heliconical 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]. Pump intensity was controlled by a half wave plate combined with a Glan-laser polarizer. 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.

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 probe beam on the sample to a waist of 300 μm.

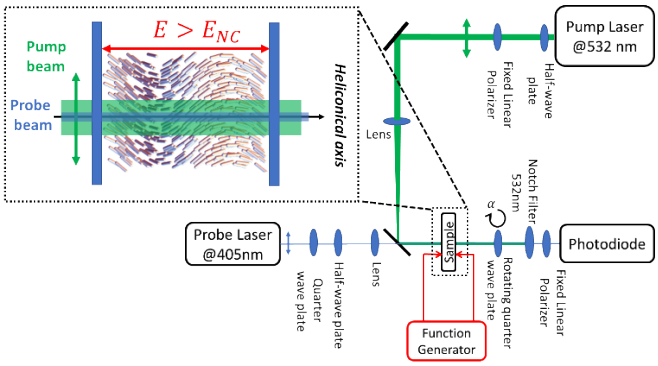

Fig.1. Sketch of the experimental apparatus for polarimetry measurements. On the top left an enlarged schematic view of the Ch_{OH} cell under the action of the low frequency electric field (Red) and of the pump beam (Green), different colors of the molecules represent different azimuthal and polar angles. The probe beam along the heliconical axis is indicated in blue.

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 the 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 that the axis of the rotating quarter waveplate forms with that of the Glan laser polarizer, whose direction is horizontal. Cell temperature was controlled by a CalcTek 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. The results of each measurement were then divided everytime by such baseline signal.

For a polarimeter based on fixed polarizer and rotating quarter waveplate, the transmission curves can be fitted by the function [12]:

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

where the fitting parameters $A, B, C,$ and D are related to the Stokes parameters describing the output polarization state [12]:

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

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

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

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

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 low frequency field on the polarization of the probe beam. Given the values of the critical field $E_{NC} =$ [5] 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:

$$E_{405} = \frac{P_0 E_{NC}}{n_{av}\lambda_P} = 2.72\ V/mm \qquad (5)$$

We thus chose to vary the electric 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 electric fields in the mentioned range. Dashed lines are best fits with eq.(3). The data are shown for different polarizations of the incoming probe beam: linear vertical (*a*), linear horizontal (*b*), circular left-handed (*c*), circular right-handed (*d*). The different color codes represent the intensity of the external electric field. It can be seen that as the electric field is lowered from 3 V/μm (red curve) the polarizations curves change accordingly as the resonant pitch is reached. For example: the linear horizontal state in fig. 2a (red curve) changes toward a circular state (yellow curve) to come back to a linear state (blue curve), as E decreases [13].

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 low frequency 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 plate with a periodicity of $\pi/2$ due to variations in the light polarization which is affected in similar ways by varying either the electric 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.

Data in Figure 2 and Eqs. (3,4) are used to find the Stokes parameters which determine the polarization state of the probe beam at the exit of the sample. These parameters allow us to reconstruct the polarization ellipses for each analyzed case [12,13]. Results are reported in Fig. 3 for both electrical (a-d) and optical tuning (e-h) of the Ch_{OH} pitch.

In both Fig. 2 and Fig. 3 the intensity of the probe beam was adapted looking for the more stable optical alignment not affected by the stirring of rotating optical elements. Therefore, the intensity of the probe beam is constant within every input polarization state (i.e. for every subfigure of Fig. 2 or rows of figures in Fig.3) but not for different input polarization states. Notice that Figures 3*e-h* are 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 = 2.75V/μm, in fair agreement with Eq. (5), while there is practically no effect on the right-handed circular polarized probe beam. At fixed low frequency 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. $\lambda_B\ (I_R)$ = 405 nm. This quite low light intensity is easily accounted for by considering that the applied electric field is just above the critical field E_{NC} for helix unwinding (5 V/μm vs 4.5 V/μm) , thus a weak optical torque is sufficient to restore the helical structure with a short pitch. The low pump intensity values also assure an efficient pitch tuning without affecting the symmetry of the single harmonic heliconical structure.

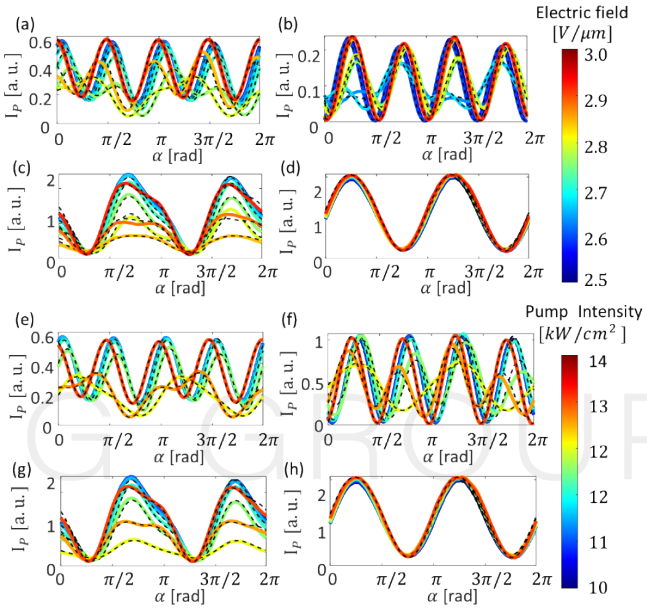

Fig.2. Polarimetric measurements of the probe beam at the exit of the Ch_{OH} sample. The upper figures (*a-d*) report the transmitted signal vs the rotation angle of the quarter wave plate for different values of the applied low frequency field (indicated by colors on the right side) with no pump beam. Dashed lines are best fits with eq.(3). 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.

The relevance of the reported observations is manifold. 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 of the low frequency 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-harmonic periodicity.

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 low frequency field. This has two important consequences: i) thanks to the efficient optical tuning of the heliconical 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 in principle 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 effect enables optically controlled phase retarders of a very simple architecture.

Even if effects of tuning of the Bragg resonance have been previously observed in some compounds including CLCs, they presented strong limitations with respect to the effect shown here. Such limitations, that make unpractical their exploitation in optical devices, are: broad and/or distorted spectra, narrow tuning range, high applied voltage, need of photochemical additive. In principle, a similar tuning could be expected in a chiral smectic C, which shows an oblique helicoidal structure similar to that of Ch_{OH}.

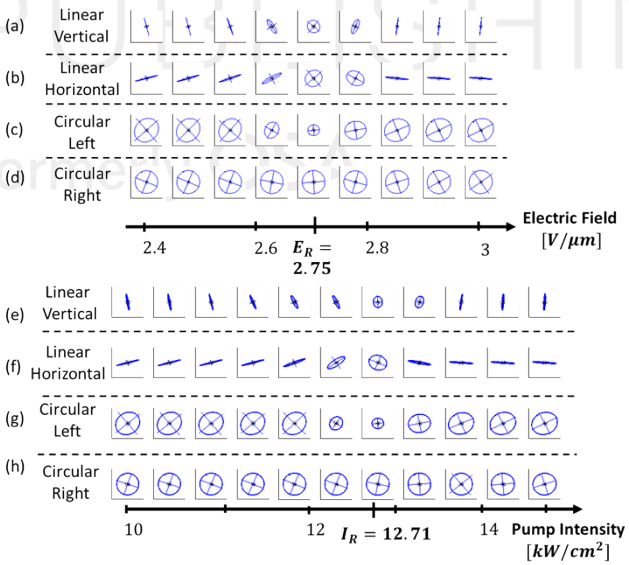

Fig.3. Plots of the polarization state of the probe beam at the exit of the Ch_{OH} sample. The upper figures (*a-d*) concern the change of the polarization state for increasing values of the applied static field with no pump beam. They correspond to different polarization of the incoming probe beam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

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 to P; these dislocations could be avoided or expelled if the cell is sufficiently thick, or if it is treated for a weak planar [14] or perpendicular director anchoring [15].

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. The tuning mechanism is rooted in the field-dependent pitch of the Ch_{OH} structure. Current polarization rotator technologies based on liquid crystal variable retarders operate at low voltages (~10V) and relatively low speed (10-100Hz). In the case of ChOH, response times are expected to be comparable with the existing values, the main advantage being the use of LC cells with very simple architecture. Indeed, ChOH cells are compact, simple in construction, do not require the use of chemical doping with absorbing dyes, allow a very wide tuning range while preserving the single mode harmonic structure and so can be ideally used with any wavelength, at least in the VIS range, and promise to be extremely cheap. Based on this, the presented study might open the door for the development of phase retarders tuned either electrically or optically.

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