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Observation of optical non reciprocity in a single layer of transparent linear chiral media with asymmetric boundaries

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We report the observation of optical non reciprocity in a material system that is very close to natural structures, such as insect skin: a single layer of linear transparent material in its ground state. The process is shown to be defined by two key parameters: the chirality of the material and its asymmetric boundary conditions. Our qualitative and preliminary quantitative analysis are in very good agreement with experimental results.

The electromagnetic non reciprocity has attracted significant attention both for fundamental (time reversal breakdown) and practical (unidirectional propagation) reasons. Apart of the traditional magnetic field excitation cases, very specific physical conditions must be satisfied by material systems to exhibit such behaviour (see Ref. [1, 2] and references therein). Thus, in one family of material systems, the use of external fields or optical nonlinearities is required [2, 3]. Multiple cells of gyroscopic media, such as cholesteric liquid crystals (CLC), containing defect layers [4] or combined with nematic liquid crystals (NLC), have been used in another family of material systems where the use of an external electric field allowed also their dynamic tuning [1]. Finally, lossy planar (two dimensional) CLC structures were also proven to exhibit optical non reciprocity [5].

In all above mentioned cases, the non-reciprocity is created artificially. We think that particularly interesting are the cases of using chiral materials, such as CLCs, which are frequently found in the nature. In fact, it is well known that a standing alone CLC layer is not changing its handedness when observed from two opposed directions and its optical properties are reciprocal. This is the reason why additional "intervention" is required [1, 4, 6]. However, in the present work, we report the observation of optical non reciprocity in a single layer of transparent (non lossy) linear CLC without using additional layers or excitation fields. This material system thus represents a situation that may be readily found in many natural systems (see later). As it will be seen later, the transmission non reciprocity here is "naturally" generated because of the asymmetric boundaries of the CLC.

Typical liquid crystal (LC) cells have symmetric geometry along the light propagation direction. Those cells are traditionally composed of two glass substrates (coated from one side by optically transparent conductive indium tin oxide (ITO) layers) that are sandwiching a thin layer of LC, with ITOs facing each other [6–8]. Usually, very thin (at the order of 50 nm) and unidirectionally rubbed polyimide (PI) layers are also added on the top of each ITO to align the LC [9]. However, some recent applications of LCs have generated the need in asymmetric cells, where the LC layer is still confined between two PI coated substrates, but only one of them bears an ITO coating [10], providing thus optically asymmetric boundary conditions for the LC (see the top center inset of the Fig. 1).

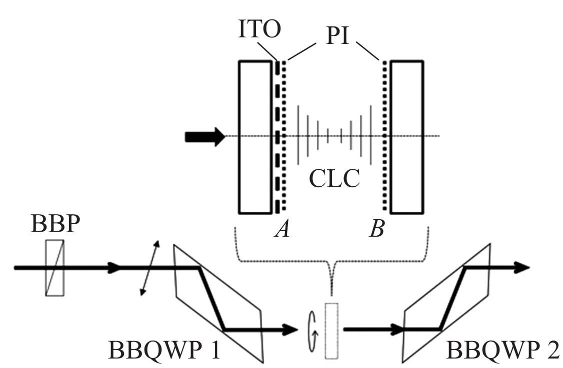

Fig. 1. The schematic geometry (bottom) of the experimental scheme used for the study of light transmission through a layer (top) of CLC with optically asymmetric boundary conditions. Top: ITO – indium tin oxide, PI – polyimide layer, CLC – cholesteric liquid crystal. *A* and *B* are light incidence directions for two sequential experiments. Bottom: BBP – broad band polarizer, BB QWP1 & 2 – broad band quarter wave plates

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We were working with such a cell of planar aligned (on both substrates) CLC that was composed of 33 wt% of CB15 and 67 wt% of MLC 2048 (both purchased from Merck). The original goal (not detailed here) of using this mixture was to create fast switchable helical structure with a spectrally selective reflection band ("bandgap") in the 1350–1600 nm region. The first component (CB15) is a right handed CLC that is usually providing an optical bandgap in the green spectral region (~520–560 nm) at room temperature. The average refractive index of CB15 was measured as $n_{av} = 1.567 \pm 0.002$ [11]. Its dielectric anisotropy is negative (tested from 100 Hz up to few 100 kHz). The second component (MLC2048) is a "dual frequency" NLC [12]. Its dielectric anisotropy is positive for "low" frequencies (~1 kHz) and is negative for "high" frequencies (~50 kHz, at room temperature).

We have used two different glass substrates to build the sandwich-like cell containing the above mentioned mixture. The first one was made from a standard microscope glass with 1 mm of thickness, while the second one was an ITO coated glass of 0.7 mm thickness (purchased from TFD). Cells were fabricated by spacing those two substrates at 8 μm distance (using spacers dispersed in the peripheral glue walls) and by using the standard method of capillary injection of the CLC mixture.

We knew [7] that the above mentioned material mixture is simultaneously sensitive to the polarization state (or angular momentum) of light as well as to its wavelength (or linear momentum). We thus started our investigations by polarization dependent transmission measurements by using Varian (Cary 500 scan) spectrophotometer. In order to obtain various polarization states (e.g., circularly polarized) of the broadband probe beam, we have built a simple setup inside the spectrometer (bottom of the Fig. 1). We used a broad band polarizer (BBP) that changed the original unpolarized beam into linear polarized one and a Fresnel rhomb, as broadband quarter wave plate (BBQWP1), which has generated the circularly polarized probe beam. The polarizer's axis' direction was tilted at 45° relative to the horizontal direction and, when needed, we could turn it to 90° to change the circularity handedness (left–right) of the probe beam (after the BBQWP1). The second Fresnel rhomb (BBQWP2) transformed the probe's polarization back to linear one and also brought its propagation axis into the original direction. The CLC cell (with parallel but dissimilar interfaces, see the top inset of the Fig. 1) was placed between two BBQWPs, in the area where the probe beam was circularly polarized.

The experimental procedure was the following: we placed the sample of CLC between two BBQWPs and we measured the transmission spectrum; then we turned the sample at 180° (with respect to the vertical axis) and we measured the spectrum again (for the opposed direction of propagation of light through the cell). We have repeated this procedure many times for various polarization states of the probe beam and for each position of the cell (the cells were also removed and placed back again multiple times to evaluate the contributions of various measurement errors).

The obtained transmission spectra of our cells clearly show non-reciprocal behavior. Indeed, as one can see, from the Fig. 2a, the transmittance of the cell in the spectral bandgap area is noticeably higher for the case when the left circularly polarized light (non-resonant with the CLC) is incident from the side B (see the top inset of Fig. 1). In fact, this non-reciprocity was observed for all key states of incident beam's polarization (including

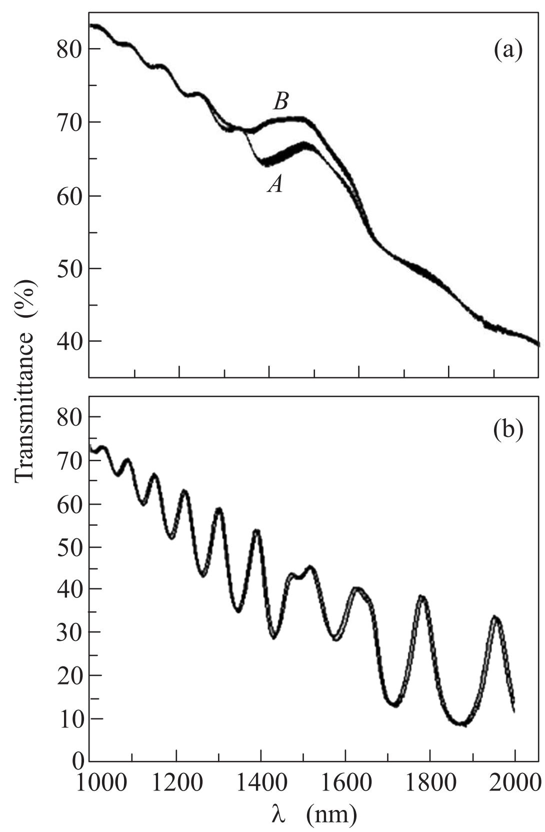

Fig. 2. Non resonant circularly polarized light transmission spectra (through the planar cell of CLC) detected for two opposed directions of light propagation through the cell. (a) – The cell was built with asymmetric optical boundary conditions. (b) – The cell was built with symmetric optical boundary conditions (see Fig. 1 for the definitions of *A* & *B* cases)

for linear parallel and linear perpendicular to rubbing direction, both circularities, etc.) but the corresponding results will be detailed in a following article (for the sake of the shortness of this communication).

The same measurement, performed with the same type of the cell of the same CLC mixture, but containing two ITO coated substrates (and thus with optically symmetric boundary conditions for the CLC) shows no such effect (Fig. 2b). This is emphasizing the key role of the asymmetry boundary conditions in the observed non-reciprocity. Note that the lower transmission and additional oscillations (in Fig. 2b) are resulted by the presence of the second ITO and the corresponding Fabry-Perrot effect.

In addition, there was no such effect of non-reciprocity for the same asymmetric cells, which was containing an NLC (non gyroscopic LC) and for all individual elements of used cells (ITO coated glass substrates, etc.). This confirms that the chirality of CLC layer also plays a major role in the observed non-reciprocal transmission effect.

Let us consider a layer of CLC with optically non-symmetrical boundary conditions to develop a simple qualitative analysis of the processes involved (Fig. 3). The interface A of the CLC represents the boundary

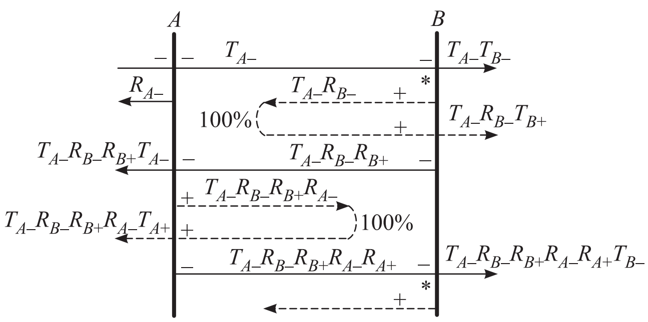

Fig. 3. Qualitative description of the physical origins of the optical non reciprocity in the layer of CLC with asymmetric optical boundary conditions A and B (light is incident on the interface A)

with the ITO, while the interface B is the boundary with the ordinary glass (both are PI coated). Given that the experimentally observed non-reciprocity was presented (Fig. 2) for the case of the probe beam having non-resonant circular polarization (designated as "$-$" in the Fig. 3), we shall consider this specific case only. Then, we can start by analyzing the probe beam's transmission for an incidence from the interface A (propagating from left to right). The coefficient T_{A_-} will represent the transmission coefficient (by intensity) of the probe (with "$-$" polarization) through the interface A. The coefficient T_{B_-} will represent the transmission coefficient of the probe (with "$-$" polarization) through the interface B. Thus, for the first passage, the probe's transmission (through the interfaces A and B) may be expressed by the "aggregate" coefficient $T_{A_-}T_{B_-}$.

Furthermore, the coefficient R_{B_-} will represent the reflection coefficient of the probe (with "$-$" polarization) from the interface B. As it is well known, the circular polarization state of light is inversed when it is reflected from a simple interface (since we have the same direction of rotation of light's electric field, but an opposed direction of its wave vector). Thus, given that the handedness of the CLC is not changed, we shall now have a resonantly polarized probe beam (designated as "$+$" in the Fig. 3), propagating from right to left, after its first reflection from the interface B. This light will be completely (100%) reflected from the bulk CLC [6] if the parameters of the CLC layer are appropriately chosen. In addition, it is also well known [6] that the polarization state of the resonantly reflected (from the CLC) light will remain the same (here "$+$"). This resonantly reflected beam will generate "another" transmitted beam in the second passage. However, given that the CLC is a gyrotropic media, the refractive indices (n_+ and n_-), seen by opposed circular polarizations ("$+$" and "$-$"), are different. Thus, the transmission and reflection coefficients of our interfaces (A and B) are not the same for two opposed circular polarizations. That is the reason why the transmission term for the second passage will be expressed by the coefficient $T_{A_-}T_{B_-}T_{B_+}$. This beam (with polarization "$+$") will be reflected from the interface B with the coefficient $T_{A_-}T_{B_-}R_{B_+}$ and also with inversion of its polarization into the state "$-$". This beam will propagate across the CLC and will be reflected from the interface A with the coefficient $T_{A_-}T_{B_-}R_{B_+}R_{A_-}$ and with inversion of its polarization state (into "$+$"), which will be then 100% reflected from the bulk CLC, etc. The further analysis of the light propagation (with another reflection from the interface A) shows that, after the third passage, the aggregate coefficient of transmission (from left to right) will be given as $T_{A_-}T_{B_-}R_{B_+}R_{A_-}R_{A_+}T_{B_-}$. We shall not continue further this consideration since it will simply repeat the same cycles (delimited by two star signs on the top right and bottom right of the Fig. 3, near the interface B), but with smaller contributions. Given the non-coherent character of the light source used, we can estimate the total transmission coefficient (for the light's incidence from the interface A) as

$$T_{A\text{-to-}B} = T_{A_-}T_{B_-} + T_{A_-}R_{B_-}T_{B_+} + \\ + T_{A_-}R_{B_-}R_{B_+}R_{A_-}R_{A_+}T_{B_-} \cdots . \quad (1)$$

Similar analyses, conducted for the light incidence from the interface B (propagating from right to left), gives us an aggregate coefficient of transmission (for first three passages) that may be presented as

$$T_{B-\text{to}-A} = T_{B_-}T_{A_-} + T_{B_-}R_{A_-}T_{A_+} + \\ + T_{B_-}R_{A_-}R_{A_+}R_{B_-}R_{B_+}T_{A_-}\cdots . \quad (2)$$

As one can see, the second and third terms of equations (1) and (2) are different. This difference disappears (confirming thus our observations) if the boundary conditions are the same

$$T_{A_\pm} = T_{B_\pm} \text{ and } R_{A_\pm} = R_{B_\pm}. \quad (3)$$

Also, if the boundary asymmetry is still present, but the circular anisotropy of the CLC is very weak

$$T_{A_-} \approx T_{A_+} = T_A, \quad R_{A_-} \approx R_{A_+} = R_A, \\ T_{B_-} \approx T_{B_+} = T_B, \quad R_{B_-} \approx R_{B_+} = R_B, \quad (4)$$

then we obtain a simple relation for the non-reciprocity

$$\frac{T_{A-\text{to}-B}}{T_{B-\text{to}-A}} \approx \frac{1 + R_B + R_B^2R_A^2}{1 + R_A + R_A^2R_B^2}. \quad (5)$$

Thus, the difference in reflections R_A and R_B is the major contributor of non-reciprocity. However, the chiral character of the CLC is necessary for the non-reciprocity since, otherwise, the travel of light in the media will be different and the ratio of transmissions in opposed directions will be expressed as

$$\frac{T_{A-\text{to}-B}}{T_{B-\text{to}-A}} = \frac{1 + R_BR_A + R_B^2R_A^2}{1 + R_AR_B + R_A^2R_B^2} = 1. \quad (6)$$

This again confirms our experimental results.

To further validate the described (in the equation (5)) hypothesis of the observed non-reciprocity, we have done additional experiments by measuring the coefficients of reflection and transmission for the microscope glass and ITO coated glass in air at normal incidence at a wavelength that was inside the spectral bandgap area (= 1481 nm) of our CLC. The obtained values are as follows:

$T_{A(\text{ITO-glass})} = 0.61 \pm 0.015$, $R_{A(\text{ITO-glass})} = 0.2 \pm \pm 0.015$, $T_{B(\text{glass})} = 0.87 \pm 0.015$, $R_{B(\text{glass})} = 0.07 \pm 0.015$. Using those values we can estimate the effective refractive index values of the ITO and glass substrates, respectively as $n_{\text{ITO}} \approx 2.0 \pm 0.05$ and $n_G \approx 1.50 \pm 0.05$. Using the average refractive index of the CLC we can then estimate the values of $R_A \approx 0.019 \pm 0.002$ and $R_B \approx 0.0005 \pm 0.0001$. Obviously those reflection coefficients are smaller compared to the values obtained in air because of the smaller refractive index mismatch in the CLC cell. In this case, the expression (5) could be further simplified to result in the following form

$$T_{A-\text{to}-B}/T_{B-\text{to}-A} \approx (1 + R_B)/(1 + R_A). \quad (7)$$

We thus obtain estimation for the non-reciprocity coefficient that should be $T_{A-\text{to}-B}/T_{B-\text{to}-A} \approx 0.98 \pm 0.09$. The same ratio, calculated from the Fig. 2a, is $\approx 0.953 \pm \pm 0.008$. We think that, given the approximations made, this is a rather good agreement. In addition, this simple formula (7) also shows that the best ratio of non-reciprocity may achieve the factor of 2, when $R_B = 1$ and $R_A = 0$.

We have used the method, described in Ref. [4], trying to reproduce the experimentally observed non-reciprocal behavior. The system discussed here can be treated as a multi-layer system: Isotropic Layer (1) – CLC Layer – Isotropic Layer (2). According to Ambartsumian's layer addition modified method, if there is a system consisting of two adjacent (from left to right) layers, A and B, then the reflection and transmission matrices of the system, $A + B$, viz. $\widehat{R}_{A+B}$ and $\widehat{T}_{A+B}$, are determined in terms of similar matrices of its component layers by the matrix equations:

$$\widehat{R}_{A+B} = \widetilde{R}_A + \widetilde{\widetilde{T}}_A\widehat{R}_B[\widetilde{I} - \widetilde{\widetilde{R}}_A\widehat{R}_B]^{-1}\widehat{T}_A, \\ \widehat{T}_{A+B} = \widehat{T}_B[\widetilde{I} - \widetilde{\widetilde{R}}_A\widehat{R}_B]^{-1}\widehat{T}_A, \quad (8)$$

where the tilde denotes the corresponding reflection and transmission matrices for the reverse direction of light propagation, and I is the unit matrix. The exact reflection and transmission matrices for a finite CLC layer (at light normal incidence) and for an Isotropic Layer are well known [13, 14]. First, we sew the Isotropic Layer (2) with the CLC Layer from the left side, using matrix Eqs. (8). In the second stage, we sew the Isotropic Layer (1) with the obtained CLC Layer – Isotropic Layer (2) system, again from the left side.

Here, we present (in Fig. 4) one particular case of simulation corresponding to our experimental results (Fig. 2a). The simulation parameters used are as follows: the effective thickness of the glass A (ITO coated glass) is considered 1 mm and its effective refractive index is described by the equation: $n_A = 1.4 + 2(\lambda - 1)/\lambda$. Ordinary and extraordinary refractive indexes of the CLC are $n_0 = 1.5$, $n_e = 1.7$ respectively. Helix pitch of the CLC is $p = 0.96\,\mu$m (the distance at which the LC molecules rotate at 360°) and the number of entire helixes is $t = 6$. Thickness of the glass B (ordinary glass) is 0.5 mm and its refractive index is 1.5. As one can see (Fig. 4), we obtain very good agreement between the experimental and theoretical results for two opposed directions of light

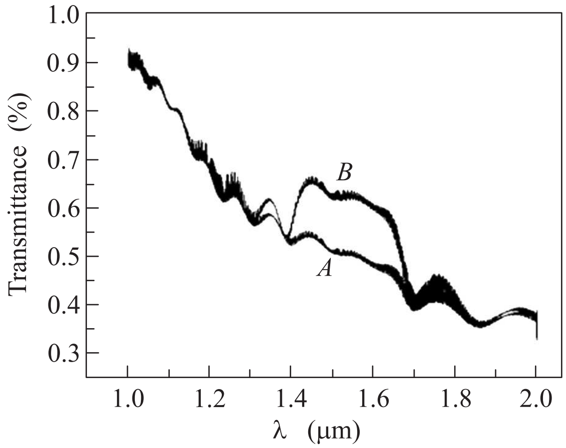

Fig. 4. Theoretical description of light transmission through the CLC cell with asymmetric boundary conditions in two opposed directions A and B (see Figs. 1 and 2a)

propagation through the CLC cell that was built with asymmetric optical boundary conditions.

We have observed and explained, in the first approximation, an optical non reciprocity phenomenon in a linear and non-absorbing material layer without using excitation fields. We have shown that this non reciprocity is defined by two key parameters of the media: the chirality of the layer and the asymmetry of its boundary conditions. We believe that the described system is very simple (well suited for potential applications) and, in the same time, is rich since it has very specific dependence upon the angular and linear momentums of light. In addition, we think that the reported non-reciprocity might also have interesting implications in the natural evolution given that such chiral material systems (with asymmetric boundary conditions) are abundantly present in the natural environment, such as various insect skins, which are chiral and have asymmetric boundaries (air and body tissue) [5, 15, 16].

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