Reflection and Transmission Properties of Light in Chiral Liquid-crystalline Photonic Structures with Pitch Gradient and the Phenomenon of Nonreciprocity

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Abstract: In frame of this paper we have investigated reflection and transmission properties of light in chiral liquid-crystalline photonic structures with inhomogeneous pitch. We have experimentally confirmed and theoretically justified the existence of the phenomenon of nonreciprocity in the mentioned above systems. The liquid-crystalline cells through the contact of two and more cholesteric liquid crystals with different pitches and chiralities were prepared. The problem was solved by Berremann 4x4 matrix method. Our results can be used in optical diodes, transistors and logical elements, as well as for expansion of Bragg's reflection range, namely in the systems as a broadband mirrors for circularly polarized light and in spatially tunable liquid-crystalline lasers with a pitch gradient.

1 INTRODUCTION

During the last decade photonic crystals and various photonic structures became one of the most interesting objects of physics due to their unique physical properties and their relevance for development of new electro-optical and photonic devices. The main peculiarity of PCs is the fact, that they exhibit photonic band gaps (PBGs), namely they prevent light propagation in certain frequency ranges, which can be suitably tuned modifying their structure (Joannopoulos et al. 1995); (Sakoda, 2001); (Johnson and Joannopoulos, 2002); (Soukoulis, 2001). Chiral photonic crystals (CPCs), namely cholesteric liquid crystals (ChLCs) are classic examples of PCs. The main difference between CPCs and usual photonic crystals lies in the fact that the photonic band gap in CPCs exists only for one polarization (in case of normal light incidence) coinciding with the chiral medium helix sign. ChLCs are particularly interesting one-dimensional materials because of their spontaneous self-assembly into periodic structures and the fact that the photonic band gap can be tuned over a broad range of frequencies. In terms of their optical properties, a prominent feature of cholesterics is the helical structure of their director axes. This helicity gives rise to selective reflection and transmission of circularly polarized light (Kolomzarov, et al., 1999); (De Gennes and Prost, 1993). As we mentioned above, the periodic structure of ChLCs leads to a polarization sensitive photonic band gap. Its location and width depend on the pitch p and the ordinary (n_e) and extraordinary (n_o) refractive indices of cholesteric material. For ChLCs the reflection band is defined by $n_o p<\lambda<n_e p$. Within the stop band only circularly polarized light that has the same handedness as the ChLC helix is selectively reflected. Light polarized with the opposite handedness and light with wavelengths outside the stop band are both transmitted through the ChLC cell. Maximum reflection efficiency at normal incidence for ambient light is 50%. Therefore, ChLC systems, that can reflect both right- and left-circularly polarized light simultaneously, attract scientists' attention. For example, M. Mitov et al. have obtained a way to exceed the 50% reflectance limit (Mitov et al., 1999); (Dimitrios et al., 2006); (Belalia et al., 2006). It is worthy to mention about nonreciprocal photonic crystals, which are also of special interest, the more so given that some new mechanisms of nonreciprocity have been proposed in recent years (Scalora et al., 1994); (Poladian, 1996). The importance of nonreciprocal photonic crystals is related, in particular, to the possibility of

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creating optical diodes, unilateral reflectors etc. Accordingly, a device transmitting light in one direction and blocking its propagation in the reverse direction (i.e., characterized by nonreciprocal transmission) may be called a purely optical diode. The importance of the creation and characterization of optical devices (optical diodes, transistors, etc.) analogous to electrical devices is related, in particular, to the general tendency of using optical signals instead of electrical. This trend is explained by some advantages and extremely broad possibilities offered by optical signals.

From applications point of view, photonic-crystal-based devices should be important for controlling of optical properties, because they do not rely on the intrinsic properties of the constituent materials. In particular, the properties of chiral photonic crystals are dependent on the boundary conditions, which can be engineered to suit a wide variety of diverse applications. So, the optical devices, constructed on the bases of chiral photonic crystals result intelligent, multifunctional and tunable optics, which possess such good traits, as their compactness, small losses, high reliability and compatibility with other devices. PCs, such as cholesteric liquid crystals, artificial chiral-made crystals etc., are used in switches, laser devices, tunable filters, tunable optical chips, nonlinear optical devices, colour displays, thermometers, optical polarizers, as well as in thermal mapping and for solar energy savings in building windows (Yeh and Gu, 1999); (Wu and Yang, 2001); (Bahadur, 1991); (Foresi et al., 1997); (Mochizuki et al., 2000); (Furumi and Sakka, 2006). Therefore, we can say that ChLCs are widely used in basic research as well as in commercial purposes.

2 DIFFUSION AND PITCH GRADIENT PECULIARITIES IN CPC STRUCTURES

The pitch change is caused as a result of the molecular diffusion of the chiral components into the ChLC as shown schematically in Fig.1. The diffusion of molecules of LC in the depth direction of the cell has been used to create a ChLC material with a pitch gradient. A diffusion process between compounds and therefore, a chirality gradient may occur in a direction perpendicular to the plane of films. Because of deformation the helical pitch simply followed the changes in sample thickness.

At the beginning of the process the two original parts can still be distinguished with their small and large periodicity, but the interface is no longer visible. Clearly the adjacent surfaces have merged and diffusion has occurred. Finally, redistribution of the chiral compound might allow the material to adjust its local equilibrium pitch. The speed of CLCs mixing by diffusion depends on four main parameters: temperature, size (mass) of the diffusing particles, viscosity of the structure and pressure. The main characteristics of diffused layers are: surface resistance or surface concentration of mixture, depth of doped layer, as well as distribution of mixture's density in that layer. The concentration gradient in the cell with the thickness d will be determined by the following equation:

$$\nabla C = (C_1 - C_2) / d, \quad (1)$$

where C_1 and C_2 are the concentrations of diffusion layers. For our sample the concentration gradient is of the order about 2.8×10^{23} sm^{-4}. So, the pitch gradient is the consequence of the concentration gradient. As we know diffusion process is characterized by diffusion constant, which is the quantitative characteristic of diffusion velocity. It should be incorporated in the model for it to be capable of predicting the pitch change phenomena for given experimental conditions. It is defined by medium's properties. The diffusion process throughout the sample allows the concentration become uniform.

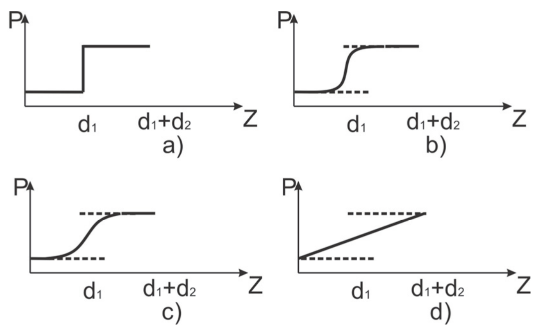

Figure 1: The model of ChLC pitch's profile: a) at the beginning of diffusion, b),c) in the middle of diffusion process and d) at the end of diffusion.

The pitch profile is represented by the following function:

$$p(z) = p_1 - \frac{2(p_2 - p_1)}{(z_2 - z_1)^3}(z - z_1)^2(z - \frac{3z_2 - z_1}{2}) , \quad (2)$$

where p_1 and p_2 are pitches of two cholesteric liquid crystals.

Although the pitch gradient is formed along the one direction, the liquid-crystalline material also diffuses flowing on the transverse plane, consequently the total thickness gradually diminishes as the thermal diffusion process lasts longer. As we know concentration increases when pitch decreases. Helical twisting power (HTP) is expressed by the pitch and concentration of ChLC:

$$HTP=1/(P*C), \quad (3)$$

According to this equation ChLC pitch (reflection band) is determined based on the concentration of ChLC.

So, the ChLC film with a lateral pitch gradient was prepared by thermally activated molecular diffusion across the interface of two films with disparate chiral concentrations. As we mentioned above, in order to broaden the selective reflection band we have changed the helix pitch. The pitch of ChLCs can be varied by adding chiral dopants, by adjusting temperature or by irradiating with light of a specific wavelength, namely the helical pitch can also be altered by UV irradiation of ChLC containing light-sensitive molecules with variable chirality controlling the pitch (Bodrovsky et al., 2004). So, many external stimuli, such as pressure, electric and magnetic fields, temperature and light can be used to modify the pitch and /or average refractive index of the CLCs (Munoz et al., 2001); (Funamoto et al., 2005); (Furumi et al., 2004); (Sackmann, 1971). The spectral position of the reflection band can be tuned throughout the visible spectrum: but since the typical birefringence of LCs in the optical region is $\Delta n=n_e-n_o$~0,15-0,2, the typical reflection bandwidth is limited to 50-100nm. This fact is insufficient for some applications. By introducing a pitch gradient in the helix (reflection bandwidth with pitch gradient CLCs is defined by $\Delta\lambda= n_e p_2- n_o p_1$) we show that reflection may occur of a wavelength bandwidth greater 300nm. Therefore the spectral position of reflection band can be controlled in whole visible spectrum. A way to make cholesteric films reflecting in a broad wavelength band consists in associating different cholesteric pitches in the same film. A controlled helix pitch modulation in the in-plane direction of a planarly aligned cholesteric liquid crystal cell by using photopolymerizable cholesteric liquid-crystals is demonstrated in the work of H.Yoshida and Masanori Ozaki et al. (Hiroyuki et al., 2008). We proposed an interesting method to produce film with pitch gradient based on a thermal processing and we studied the optical properties of this film with respect to the time of processing. Let us recall that our intention was to make double- and multiple-pitched specimen. The type of linear pitch gradient for achieving a broadband reflection was analysed and the number of chiral pitches required for establishing the stop-band was simulated. In this case we have to know answers of two important questions, namely: how to adjust the pitch and how to keep it constant when the right value is reached? It is worthy to mention, that we considered the helical pitch variation profile uniformly along the two directions. This idealization has been proven far from being valid in practice: the cholesteric films after the heating procedure showed both transversal variations and additionally, a disturbed longitudinal profile.

3 EXPERIMENT

3.1 Sample Preparation

For investigation of spectral properties of ChLCs, we have used right-handed pelargonium, left-handed oleate and E7 nematic liquid crystals. We have prepared cholesteric liquid-crystalline cell through the contact of two and more cholesteric liquid crystals with different pitches and chiralities. The inner surfaces of glass substrates were first coated with thin polymide layer and then they were uniaxially rubbed with special material. As a result, the orientation of CLCs director was parallel to the surfaces, which means that the helix axis was perpendicular to the surfaces of the cell. The substrates were coated with mixtures of different pitches and chiralities. The mixture was drop-filled into the empty sandwiched cell and it was sealed with glue. After this procedure the cell showed the pitch gradient produced by the mixing process. Polyethylene films with a thickness of 15μm were used as the cell spacers and the distance between them was 3,5mm.

3.2 Experimental Set-up

In order to investigate the transmission and reflection spectra for unpolarized, linear and circular polarized lights (at normal incidence) we have assembled the experimental set-up, depicted in Figure 2. In our experiment StellarNet spectrometer with optical resolution of 0,75 nm was used.

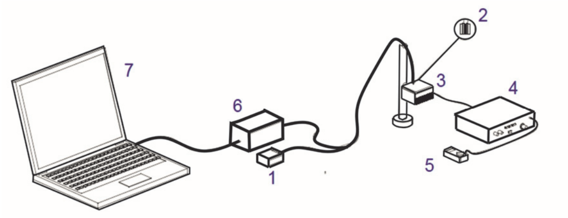

Figure 2: Scheme of experimental set-up for investigation of discussed system's reflection and transmission spectra: 1. Tungsten-krypton lamp. 2. CLC cell, 3. Microrefrigerator, 4. Controller of temperature, 5.Tester, 6. Spectrometer, 7. PC.

4 RESULTS AND DISCUSSION

It is worthy to mention about phenomenon of nonreciprocity as well. It is in the centre of attention of scientists, who are interested in the questions of reflection and transmission.

Its experimental and theoretical justifications were given for different liquid-crystalline systems (Manipatruni et al., 2009); (Vardanyan and Gevorgyan, 2005); (Miroshnichenko et al., 2010), but in chiral photonic liquid-crystalline structures with inhomogeneous pitch it isn't justified yet, so experimental and theoretical justifications of this important phenomenon is considered one of the basic questions of this paper. The meaning of nonreciprocity phenomenon is that the optical properties of liquid-crystalline system are different from two different sides of the system, which means that reflections of light from different sides of the system are different. For our system the absorption can be neglected, so for transmission coefficients the phenomenon of nonreciprocity also takes place. The phenomenon of nonreciprocity is widely used in optical diodes, transistors, as well as in different logical elements.

Figure 3 shows the reflection spectra of the cell for different moments of diffusion process. At the beginning of the diffusion process two peaks are separated obviously, then two peaks come to close each other. Finally, at the end of diffusion process these two peaks almost can't be distinguished.

5 METHOD OF ANALYSIS

In order to simulate our experimental results, we use Berremann 4x4 matrix method. There are many methods of solving the problem of electromagnetic wave propagation through a 1D anisotropic PC layer

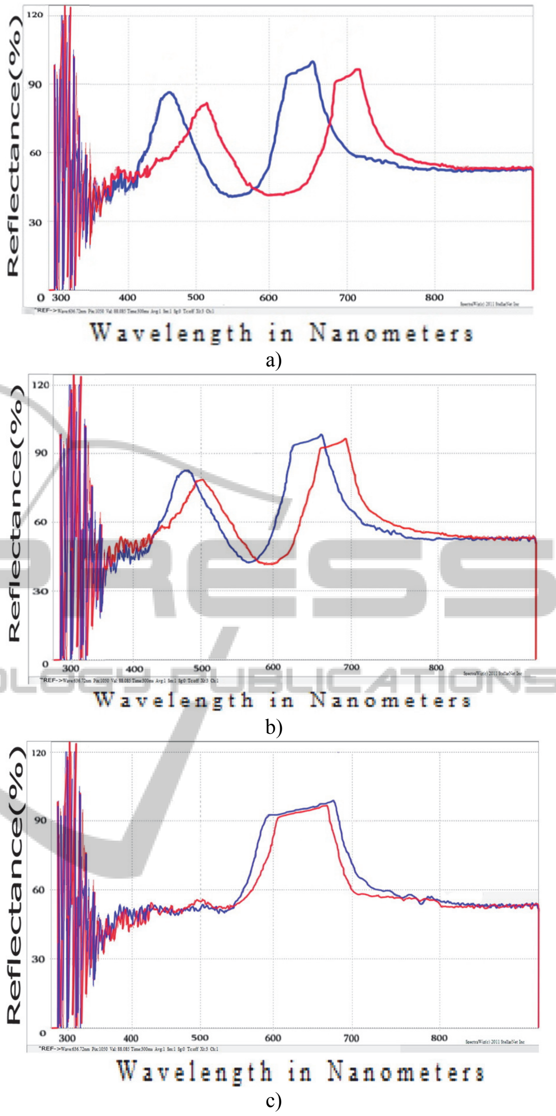

Figure 3: Reflection spectra of the cell for different moments of diffusion process: a)at the beginning of diffusion, b) in the middle of diffusion process and c)at the end of diffusion.

but it seems that the most applicable one has recently become the Berremann's method (Berreman, 1972). According to Berreman's method, the Maxwell's equations will presented as follows:

$$\frac{\partial}{\partial z}\vec{\psi} = i\frac{\omega}{c}\Delta\vec{\psi}\,, \tag{4}$$

where $\vec{\psi} = \left[E_x, H_y, E_y, -H_x\right]^T$ is the field generalized vector p Δ is the well-known

Berremann's 4x4matrix.

The solution of equation (4) can be expressed by the characteristic matrix $\hat{P}$:

$$\psi(z_2) = P(z_2, z_1)\psi(z_2) \qquad (5)$$

For the multilayer structures $\hat{P}$ is defined by the following product:

$$\hat{P} = \hat{P}^{(n)} \cdot \hat{P}^{(n-1)} \cdot \ldots \cdot \hat{P}^{(3)} \cdot \hat{P}^{(1)} \cdot \hat{P}^{(1)} \qquad (6)$$

In Figure 4 simulation results are presented, namely the reflection spectra of the cell are calculated at the beginning and at the end of diffusion process.

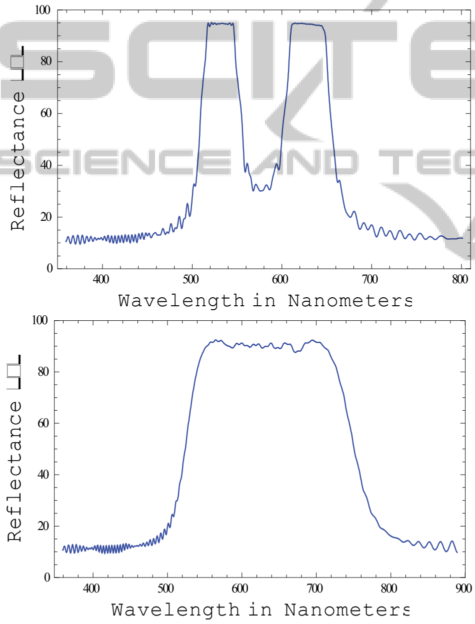

Figure 4: Reflection dependences on wavelength of the cell for different moments of diffusion process.

As it is obvious from the graphs, the simulation results agree with experiment. Let us not that the absorption of the ChLC medium is neglected.

6 CONCLUSIONS

In summary, we theoretically designed the ChLC cell with two and more cholesterics of different pitches and chiralities, as well as experimentally demonstrated the peculiarities of this system. Thanks to the diffusion process, we created a gradient in the pitch of the helix. The pitch gradient was stable for few months. So, as a result of the sensitivity of the helical pitch we create a new material. We also showed that the bandwidth of cholesteric reflection was broaden. Our studies provide important insights into self-assembled photonic investigations to go into details about the diffusion mechanism between the individual films inside the cell. The comparison of experimental results with the theoretical predictions confirms the validity of the approach. Work is in progress aiming to improve our sistem's nonreciprocity.

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REFERENCES

Joannopoulos, J., Meade, R., and Winn, J., 1995. *Photonic Crystals*, Princeton Univ.

Sakoda, K., 2001. *Optical properties of Photonic crystals*, Berlin: Springer.

Johnson, S., G., and Joannopoulos, J., 2002. Photonic Crystals: The Road from Theory to Practice, Boston: Kluwer.

Soukoulis, C., M., (Ed), 2002. *Photonic Crystals and Light Localization in the 21st Century*. NATO Science Series C: Vol.563. 616 p.

Kolomzarov, Yu., Kozachenko, A., Lev, B., Nazarenko, V., and Sorokin, V., 1999. *Some peculiarities of angular selective reflection of cholesteric liquid crystals in an electric field*, Jpn. J. Appl. Phys., 814-817, Part1, No2A, February.

De Gennes, P., G., Prost, J., 1993. *The Physics of Liquid Crystals*, Clarendon Press, Oxford, 271-280.

Mitov, M., Boudet, A., and Sopena, A., 1999. *From selective to wind-band light reflection: a simple thermal diffusion in a glassy cholesteric liquid crystal*, Eur.Phys.J.B8, 327-330.

Dimitrios C., Emmanouil E., Kriezis, Michel Mitov, Corinne Binet, 2006. *Theoretical and experimental optical studies of cholesteric liquid crystal films with thermally induced pitch gradient*, Physical Review E 73, 061701

Belalia, M., Mitov, M., Bourgeratte, C., Krallafa, A., Belhakem M., and Bormann,D., 2006. *Cholesteric liquid crystals with a helical pitch gradient: Spatial distribution of the concentration of chiral groups by Raman mapping in relation with the optical response and the microstructure*, Phys. Rev. E 74, 051704.

Scalora, M., J., P., Dowling, C., M., Bowden, and M., J., Bloemer, 1994, *The Photonic Band Edge Optical Diode*, J.Appl.Phys. 76, 2023.

Poladian, L.,1996, *Resonance mode expansions and exact*

solutions for nonuniform gratings, Phys. Rev. E 54, 2963
Yeh, P., and Gu, C., 1999. *Optics of Liquid Crystal Displays*, New York: Wiley.
Wu, S., T., and Yang, D., K., 2001. *Reflective Liquid Crystal Displays* (New York: Wiley).
Bahadur, B., 1991. Liquid Crystals: Applications and Uses, edited by (World scientific, Singapore).
Foresi, J., S., Villeneuve, P.,R., Ferrera, J., Thoen, E., R., Steinmeyer, G., Fan, S., Joannopoulos, J., D., Kimerling, L., C., Smith, H., I., and Ippen, E.,P., 1997. *Photonic-bandgap microcavities in optical waveguides*, Nature (London) 390, 143.
Mochizuki, H., Hasui, T., Shiono, T., Ikeda, T., Adachi, C., Taniguchi, Y., and Shirota, Y., 2000. *Emission Behavior of Molecularly-Doped Electroluminescent Device Using Liquid-Crystalline Matrix*, Appl. Phys. Lett. 77, 1587.
Furumi, S., and Sakka,Y., 2006. Adv. Mater. (Weinheim Ger)18, 775.
Bodrovsky, A., Boiko, N., Shibaev, V., Wendorff, J., 2004. *Photoinduced textural and optical changes in a cholesteric copolymer with azobeneze-containing side groups*, Liquid Crystals 31, 351-359.
Munoz, A., Palffy-Muhoray, F., P., and Taheri, B., 2001 *Ultraviolet lasing in cholesteric liquid crystals*, Opt. Lett. 26, 804-806.
Funamoto, K., Ozaki, M., and Yoshino, K., 2005. *Discontinuous Shift of Lasing Wavelength with Temperature in Cholesteric Liquid Crystal,* Jpn.J.Appl.Phys.42, L1523-L1525.
Furumi, S., Yokoyama, S., Otomo, A., and Mashiko, S., 2004.*Phototunable photonic bandgap in a chiral liquid crystal laser device,* Appl.Phys.Lett.84, 2491-2493.
Sackmann, E., 1971. *Photochemically induced reversible color changes in cholesteric liquid crystal*, J.Am.Chem. Soc. 93, 7088-7090.
Hiroyuki, Yoshida, Yusuke, Miura, Kazuki, Tokuoka, Satoshi Suzuki, Akihiko Fujii and Masanori Ozaki, 2008. *In-plane pitch control of cholesteric liquid crystals by formation of artificial domains via patterned photopolymerization*, 10 November , Vol. 16, No. 23, OPTICS EXPRESS 19033.
Manipatruni, S., Robinson, J.,T., and Lipson, M. 2009. *Optical Nonreciprocity in Optomechanical Structures*, Phys. Rev. Lett. 102, 213903.
Vardanyan, G., A., and Gevorgyan, A., A., 2005. *Wave nonreciprocity in inhomogeneous gyrotropic media and multilayer systems: I. Inhomogeneous gyrotropic media*, Optics and Spectroscopy Volume 99, Number 6.
Miroshnichenko, A., E., Brasellet E., and Kishvar, Y., S., 2010. *Reversible optical nonreciprocity in periodic structures with Liquid crystals*, Applied Physics Letters 96,063302.
Berreman. J., D., W., 1972. Opt. Soc. Am., 62, 502-510.