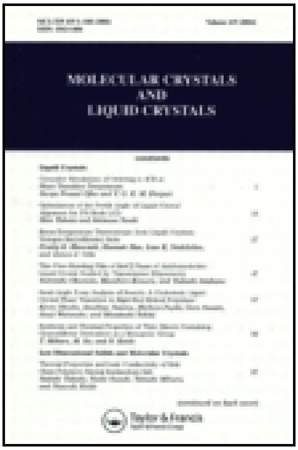

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Photonic Band-Gap Modeling of Cholesteric Liquid Crystals with Periodic Pitch Modulations

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The authors model a cholesteric liquid crystal material in which periodic elongation or shortening of the pitch has been introduced, and investigate its optical properties by the plane wave expansion method. The calculated photonic band structure revealed a continuous shift of the selective reflection band depending on the sign and degree of the modulation in pitch introduced. Anomalous selective reflection band-gaps were also observed at frequencies higher and lower than the original band-gap of the unmodified cholesteric liquid crystal, due to the longer periodicity experienced by the light interacting with the material.

Keywords: cholesteric liquid crystal; photonic band-gap material; structural defect

INTRODUCTION

Periodic dielectric media, or more commonly known as photonic crystals (PhC) possess attractive optical properties which allows one to control light propagating through the material [1]. One can observe a photonic band-gap where light within a certain frequency range is prohibited to transmit, depending on the period, refractive index and the order of periodicity. While a photonic band-gap exhibited in perfect dielectric lattices is in itself a fascinating phenomenon which allows

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one to realize perfect mirrors or gain enhancement at the band-edge frequency, one trend in the PhC field has been to add functionality into these materials by intentionally breaking the lattice. Introduction of a structural defect is known to cause a defect mode or a localized state to appear in the usually non-transmitting band-gap [2], thus allowing one to realize photonic devices such as narrow-band-pass filters [3], low-threshold lasers [4] or waveguides [5].

Liquid crystal containing PhCs have recently attracted considerable attention, mostly in view of applications [6]. Photonic devices should be tunable so that certain specifications can be met for various applications, and liquid crystals allow them to be tunable, by means of external stimuli [7]. Both experimental and theoretical reports have been made on composite PhCs with nematic liquid crystals, which allow the optical properties to be tuned by causing a reorientation of the liquid crystalline molecules by an electrical field [8–10].

From such a perspective, cholesteric liquid crystals (ChLCs) can be viewed as candidate materials for PhCs, since they spontaneously form a periodic lattice. The chiral constituents cause the material to self-organize into a helicoidal structure, which owing to the birefringence of the liquid crystal forms a periodic refractive index modulation along the helical axis. Upon normal incidence along the helical axis, ChLCs are known to exhibit a photonic band-gap over a frequency region given by $\omega = 2\pi c/n_e p \sim 2\pi c/n_o p$, where n_e and n_o are the extraordinary and ordinary refractive indices of the liquid crysal, p is the pitch and c is the velocity of light [11]. The photonic band-gap exhibited by ChLCs possesses certain peculiarities since the structure is helically periodic: it is circularly polarization selective that only light with the same handedness as the material itself is reflected, only a single band-gap is exhibited throughout the whole spectrum unlike inorganic one-dimensional structures and the size of the band-gap is larger than that exhibited in inorganic one-dimensional structures with the same refractive indices and period. Also being a liquid crystal state, they do not require a composite system and are well suited for tuning by, for example heating or light irradiation. Numerous reports on ChLC dye lasers have been made, exploiting their PhC properties [12–14].

Defect engineering in ChLCs has recently been an active topic, with both theoretical proposals and experimental demonstrations. Yang *et al*. theoretically showed that defect modes do emerge within the photonic band-gap of ChLCs upon introducing an isotropic defect layer in ChLCs [15], while an introduction of an anisotropic defect layer was found to cause interesting effects as an optical diode [16]. A discontinuous phase jump of the director rotation was theoretically predicted as a defect characteristic to the helical structure [17], with supporting

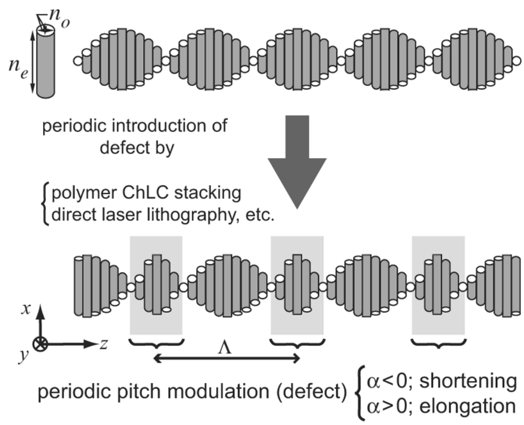

FIGURE 1 Schematic illustration of the ChLC with periodic pitch modulations.

experimental demonstration of the twist defect mode and low threshold lasing [18,19]. Tunable single defect-modes were realized in ChLCs with a locally modulated pitch introduced as the defect layer [20,21]. Utilization of a ChLC material as a defect is interesting and useful in that the defect mode wavelength is tuned as a result of varying the contrast of the pitch lengths between the bulk material and the defect.

The present study will also be on defect engineering in ChLCs, but will provide novel results since the effects of introducing multiple structural defects will be considered. In most experimental reports on defect engineering in ChLCs, polymeric ChLC materials are used to fabricate the structures, either by the stacking method or the direct laser-lithography method. These methods can easily be expanded into larger scales so that ChLCs with several or multiple defects are fabricated. Therefore it is important to understand theoretically what will happen in such complex structures, and what kind of optical properties one can expect. We focus particularly on a structure where multiple pitch-modulated regions are introduced in the bulk ChLC medium, as shown in Figure 1. Pitch modulated regions are particularly interesting as defects since in the single-defect case, the optical properties were tuned simply by varying the contrast of the pitch at the defect and the bulk. Similar results may be expected also in the case with multiple defects.

MODEL AND CALCULATION METHOD

ChLCs locally have a nematic director ordering which rotates periodically along a particular helical axis, which we set as the z-axis. For the uniform ChLC in which we assume a 0 degree tilt, the dielectric tensor distribution is described by the expression

$$\vec{\varepsilon}(z) = \begin{bmatrix} \varepsilon_{11} & \varepsilon_{12} & \varepsilon_{13} \\ \varepsilon_{21} & \varepsilon_{22} & \varepsilon_{23} \\ \varepsilon_{31} & \varepsilon_{32} & \varepsilon_{33} \end{bmatrix} = \begin{bmatrix} \varepsilon_2 \sin^2\phi + \varepsilon_1 \cos^2\phi & (\varepsilon_1 - \varepsilon_2)\cos\phi \sin\phi & 0 \\ (\varepsilon_1 - \varepsilon_2)\cos\phi\sin\phi & \varepsilon_2\cos^2\phi + \varepsilon_1 \sin^2\phi & 0 \\ 0 & 0 & \varepsilon_3 \end{bmatrix} \tag{1}$$

where ε_1, ε_2 and ε_3 are the x, y and z components of the diagonalized dielectric constant given by $\varepsilon_1 = n_e^2$ and ε_2, $\varepsilon_3 = n_o^2$. $\phi(z)$ is the in-plane angle of rotation along the helical axis, which in the case of uniform ChLCs is given by $\phi(z) = 2\pi z/p_0$, where p_0 is the pitch of the helix.

Upon modifying the structure, we assume that a helix with either a shortened or elongated pitch with respect to the initial pitch by a pitch modulation factor α, defined by $\alpha = (p_d - p_0)/p_0$ (where p_d is the modulated pitch) is introduced at a certain period Λ. For simplicity, we assume that the pitch modulated region contains one 2π rotation of the director: therefore we obtain a periodic pitch distribution

$$p(z) = p(z+a) = \begin{cases} p_d & \left(|z| < \frac{p_d}{2}\right) \\ p_0 & \left(\frac{p_d}{2} < |z| < \frac{a}{2}\right) \end{cases} \tag{2}$$

throughout the material, where $a = (\Lambda + p_d)$ is the size of the periodic lattice. The dielectric tensor of the modulated structure is still given by equation (1), though now the in-plane angle of the director rotation is given by $\phi(z) = 2\pi/p(\mathrm{z})$ and so the tensor is translation symmetric $\vec{\varepsilon}(z) = \vec{\varepsilon}(z+a)$. Figure 2 shows an example of the ε_{xx} dielectric tensor component and the pitch distribution of the assumed ChLC material with a right-handed helix, $n_e = 1.7$, $n_o = 1.5$ with a pitch modulation of factor $\alpha = -0.3$ introduced at different periods, $\Lambda = p_0$, $2p_0$, $3p_0$, $4p_0$. Note that when the modulated pitch is shorter with respect to the bulk (unmodified) ChLC, the value of α is negative. The degree of pitch modulation was varied from $-0.8 < \alpha < 0.8$ for each structure and the photonic band-structure was numerically calculated by the procedure described below.

The photonic band structure provides the dispersion relationship between the frequency and the wave number of a plane wave propagating through a medium, and is well suited to investigate the optical properties of materials possessing a certain periodic structure. The photonic band structure of the pitch modulated ChLC is determined by solving the wave equation satisfied by the magnetic field of light propagating in the medium

$$\nabla \times [\vec{\varepsilon}^{\,-1}(z) \nabla \times \vec{H}(z)] = \frac{\omega^2}{c^2}\vec{H}(z), \tag{3}$$

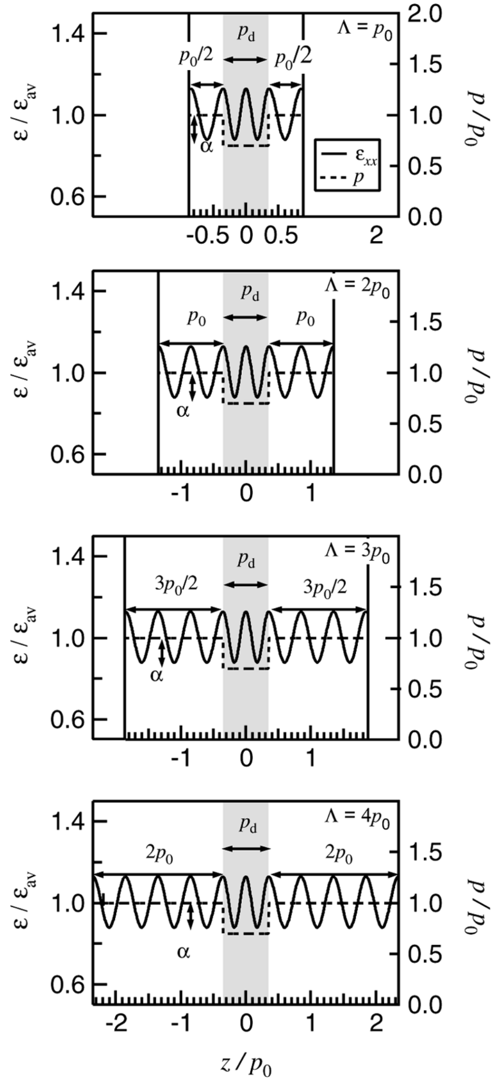

FIGURE 2 Example of the unit cells of the modeled ChLC structures with periodic pitch modulations. In the figure, the case for $\alpha = -0.3$ is shown.

where $\vec{\varepsilon}^{\,-1}(z)$ is the inverse of the dielectric tensor, and ω is the eigenfrequency.

The periodic dielectric tensor can be expanded in a Fourier series on the inverse lattice $\boldsymbol{G}$ corresponding to the period a in real space:

$$\vec{\varepsilon}^{\,-1}(z) = \sum_G \vec{\varepsilon}^{\,-1}(G)\exp(iGz); \quad G = \frac{2\pi m}{a}. \tag{4}$$

The magnetic field of light propagating in the direction of periodicity (the z-axis) can also be expanded in terms of plane waves using the Bloch-Floquet theorem,

$$\vec{H}(z) = \sum_G (h_G^x \vec{e}_x + h_G^y \vec{e}_y) \exp\{i(k+G)z\}; \quad \vec{H}(z+a) = \exp\{ika\}\vec{H}(z), \tag{5}$$

where $\vec{e}_{x,y}$ is the polarization unit vector in the x and y planes and $h_G^{x,y}$ are the respective amplitudes. Equations (3)–(5) can be converged and expressed in a matrix configuration

$$\sum_{\mathbf{G}'} \vec{\varepsilon}^{\,-1}(G-G')|k+G||k+G'| \times \begin{bmatrix} \vec{e}_y \cdot \vec{e}_y & -\vec{e}_y \cdot \vec{e}_x \\ -\vec{e}_x \cdot \vec{e}_y & \vec{e}_x \cdot \vec{e}_x \end{bmatrix} \begin{bmatrix} h_{\mathbf{G}'}^x \\ h_{\mathbf{G}'}^y \end{bmatrix} = \frac{\omega^2}{c^2} \begin{bmatrix} h_{\mathbf{G}}^x \\ h_{\mathbf{G}}^y \end{bmatrix}, \tag{6}$$

which becomes an eigenproblem that can be solved numerically. Upon computation, we employed the numerical method proposed by Ho, Chan and Soukoulis [22] which takes the inverse of the Fourier coefficients of the dielectric tensor instead of first calculating the inverse of the real space tensor and then calculating its Fourier coefficients. The number of plane waves used; i.e., the number of finite Fourier coefficients considered in our study was varied from 616 to 1160 waves, depending on the size of the periodic lattice. Although this method was originally performed on isotropic periodic media, Busch and John showed that this method was effective for anisotropic materials with an eigenfrequency convergence of 0.01% upon using 531 plane waves [6].

RESULTS AND DISCUSSION

Figure 3 shows the photonic band structure of the periodically helix-modulated ChLC structures when a pitch-shortening modulation of $\alpha = -0.3$ is introduced. The photonic band-structure of the non-modulated ChLC is also shown as a reference. The solid lines and the dashed lines indicate the eigenmodes which are either affected or unaffected by the helical structure of the ChLC, respectively. It should be noted that a photonic band-gap shown shaded in the figure is experienced only by a single eigenmode, which is in fact a elliptically polarized mode with the same circular handedness as the material itself. i.e. a selective reflection band is exhibited. The photonic band structures of the helix-modulated ChLCs show a very different optical property from that of the non-modulated ChLC. Numerous selective

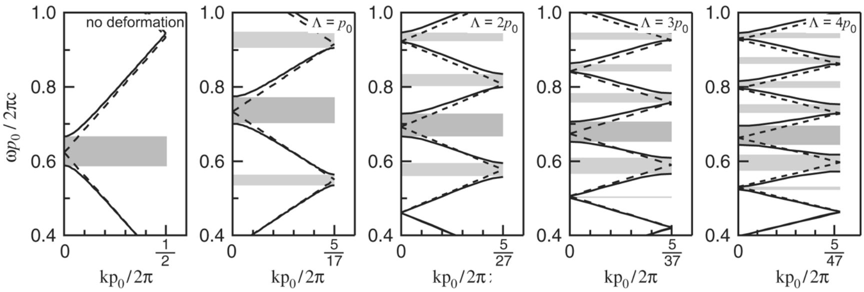

FIGURE 3 The photonic band structure of the non-modulated ChLC and the helix-modulated ChLCs with $\alpha = -0.3$. The shaded regions indicate the selective reflection band-gaps.

reflection band-gaps are observed in the periodically helix-modulated ChLC, while the non-modulated ChLC exhibits a single selective reflection band from $\omega p_0/2\pi c = 0.577 \sim 0.666$. The number of the selective reflection band-gaps is seen to increase as the period of pitch modulation is increased. For the case where $\Lambda = p_0$, 3 band-gaps appear, and for cases where $\Lambda = 2p_0$, $3p_0$ and $4p_0$, the respective band-gaps that appear are 4, 5 and 7 within the displayed frequency range of $\omega p_0/2\pi c = 0.4 \sim 1.0$. The origin of these secondary band-gaps can be understood from the dispersion relation as follows. Since the size of the unit cell, $a = (\Lambda + p_d)$ is different for each helix-deformed structure, the size of the irreducible brillouin zone, given by $k(\mathrm{X}) = \pi/a$ is also different. The secondary selective reflection band-gaps observed at the brillouin zone edges of each helix-modulated ChLC structure indicates that the secondary band-gaps originate from light which experiences the period of the unit cell for each structure: in other words, they are a result of light interacting with the periodicity of the modulation of the helical pitch, rather than the helical structure of the ChLC itself. The opening of multiple band-gaps in ChLCs has been reported in ChLC elastomers with strain-induced deformations [23]. We show that numerous band-gaps can be opened not only when the helix is deformed along the helical axis, but when a ChLC with a discretely different pitch is introduced periodically within the material. The physical origin of the secondary band-gaps; i.e., the contribution of the period of pitch modulation is also made clear by our calculations.

In each photonic band structure, there exists a wide band-gap with a width similar to the width of the band-gap of the non-modulated ChLC, shaded in dark gray in Figure 3. The spectral position of the wideband-gap shifts further away from the original selective reflection band-gap of the non-modulated ChLC as Λ is decreased, that is, as more deformations are introduced. The origin of these wide band-gaps can be attributed to the interaction of the incident light with the helical structure of the ChLC molecules. The wide reflection band-gaps appear at values of $kp_0/2\pi = 1.18$, 1.11, 1.08 and 1.06 respectively for $\Lambda = p_0 \sim 4p_0$, which is close to 1.0, where light is subject to strong interaction with the helical structure in an non-modulated ChLC. Because of the perturbations introduced in the helix-modulated ChLCs, light with $kp_0/2\pi = 1.0$ does not fulfill the Bragg condition to interact strongly with the material, but does so at a close value of k. The wide band-gaps are therefore a result of light experiencing the helical structure of the ChLC molecules in addition to the periodic modulation of the helical pitch, and therefore appear in all helix-modulated ChLC structures. The relationship between the spectral position of the wide band-gaps and the intervals of pitch modulation is explained in terms the contribution of the pitch modulated regions on the photonic band structure. For small Λ, the deformations are introduced more frequently, which means the effect of the pitch-modulated region becomes larger compared to the case with a large Λ. For the cased considered in Figure 3, $\alpha = -0.3$ is a pitch shortening deformation, which causes the band-gap to appear at higher frequency regions. Therefore the numerous $\alpha = -0.3$ modulated regions in the structure acts to shift the band-gap frequency higher, and the amount of shift becomes greater for structures with a smaller Λ.

Next, we consider the effect of α on the photonic band structure of the helix-modulated ChLCs. Figure 4 shows the frequencies of the selective reflection band-gaps as a function of α, for each helix modulated ChLC with $\Lambda = p_0 \sim 4p_0$. The two bands both above and below

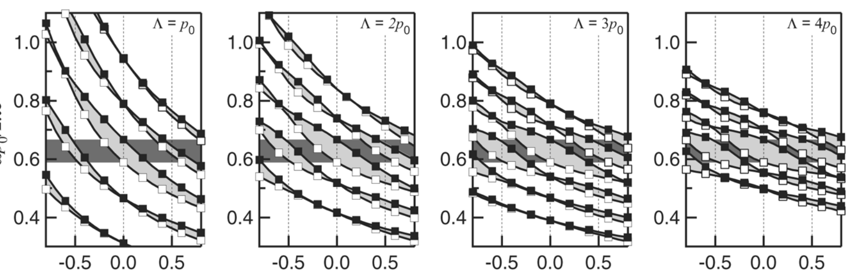

FIGURE 4 The selective reflection band-gap frequencies of the helix-modulated ChLCs as a function of varying α. The lightly shaded regions indicate the band-gaps exhibited by the structure, and the dark shaded region indicates the position of the band-gap in a non-modulated ChLC with $\alpha = 0.0$.

the initial selective reflection frequency appearing between $\omega p_0/2\pi c = 0.588 - 0.667$ are shown for each case. In all four cases of Λ, the secondary band-gaps seem to appear without a threshold of α. From the discussions above, this is because light propagating through the material experiences a periodicity of the modulating helical structure once a periodic deformation of any pitch modulation is introduced. All selective reflection band-gaps show a clear dependence on the value of α; the band-gaps shift to the higher frequency region with decreasing α, while they shift to the opposite direction with increasing α. This means that a continuous tuning of the multiple selective reflection band-gaps can be achieved by changing the degree of helix modulation. As discussed above, the amount of shift of the band-gaps is different for structures with different pitch modulation intervals. The amount of shift is larger in cases where Λ is smaller, because the contribution of the pitch-modulated regions on the optical properties is larger in cases with a small Λ.

CONCLUSION

In conclusion, we investigated the optical characteristics exhibited by ChLCs with periodic pitch modulations introduced at different intervals. Periodic deformations in the helical structure lead to the formation of secondary selective reflection bands, in which the spectral positions could be tuned either by changing the pitch modulation factor of the deformations, or the interval of pitch modulation. The number of the secondary band-gaps was found to depend largely on the interval of pitch modulation, which determines the secondary periodicity experienced by the light propagating through the structure. Understanding the physical origin of the anomalous optical properties in these complex structures should be useful for designing ChLC filters requiring specific reflective properties.

REFERENCES

[1] Joannopoulos, J. D., Meade, R. D., & Winn, J. N. (1995). *Photonic Crystals: Molding the Flow of Light*, Princeton University Press: New Jersey.
[2] Yablonovitch, E., Gmitter, T. J., Meade, R. D., Rappe, A. M., Brommer, K. D., & Joannopoulos, J. D. (1991). *Phys. Rev. Lett.*, *67*, 3380.
[3] Foresi, J. S., *et al*. (1997). *Nature* (London), *390*, 143.
[4] Painter, O., *et al*. (1999). *Science*, *284*, 1819.
[5] Mekis, A., Chen, J. C., Kurland, I., Fan, S., Villeneuve, P. R., & Joannopoulos, J. D. (1996). *Phys. Rev. Lett.*, *77*, 3787.
[6] Busch, K. & John, S. (1999). *Phys. Rev. Lett.*, *83*, 967.
[7] de Gennes, P. G. & Prost, J. (1993). *The Physics of Liquid Crystals*, Oxford University Press: Oxford, UK.

[8] Ozaki, R., Matsui, T., Ozaki, M., & Yoshino, K. (2002). *Jpn. J. Appl. Phys.*, *41*, L1482.
[9] Ozaki, R., Matsui, T., Ozaki, M., & Yoshino, K. (2003). *Appl. Phys. Lett.*, *82*, 3593.
[10] Ozaki, M., Shimoda, Y., Kasano, M., & Yoshino, K. (2002). *Adv. Mater.*, *14*, 514.
[11] Dreher, R. & Meier, G. (1973). *Phys. Rev. A*, *8*, 1616.
[12] Kopp, V. I., Fan, B., Vithana, H. K. M., & Genack, A. Z. (1998). *Opt. Lett.*, *23*, 1707.
[13] Matsui, T., Ozaki, R., Funamoto, K., Ozaki, M., & Yoshino, K. (2002). *Appl. Phys. Lett.*, *81*, 3741.
[14] Funamoto, K., Ozaki, M., & Yoshino, K. (2003). *Jpn. J. Appl. Phys.*, *42*, L1523.
[15] Yang, Y.-C., *et al*. (1999). *Phys. Rev. E*, *60*, 6852.
[16] Hwang, J., Song, M.-H., Park, B., Nishimura, S., Toyooka, T., Wu, J. W., Takanishi, Y., Ishikawa, K., & Takezoe, H. (2005). *Nat. Mater.*, *4*, 383.
[17] Kopp, V. I. & Genack, A. Z. (2002). *Phys. Rev. Lett.*, *89*, 033901.
[18] Ozaki, M., Ozaki, R., Matsui, T., & Yoshino, K. (2003). *Jpn. J. Appl. Phys.*, *42*, 472.
[19] Schmidke, J., Stille, W., & Finkelmann, H. (2003). *Phys. Rev. Lett.*, *90*, 083902.
[20] Yoshida, H., Lee, C.-H., Fujii, A., & Ozaki, M. (2006). *Appl. Phys. Lett.*, *89*, 231913.
[21] Yoshida, H., Lee, C.-H., Miura, Y., Fujii, A., & Ozaki, M. (2007). *Appl. Phys. Lett.*, *90*, 071107.
[22] Ho, K. M., Chan, C. T., & Soukoulis, C. M. (1990). *Phys. Rev. Lett.*, *65*, 3152.
[23] Bermel, P. A. & Warner, M. (2002). *Phys. Rev. E*, *65*, 056614.