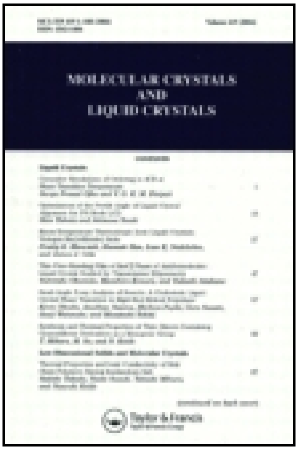

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Optical Properties of Cholesteric Liquid Crystals with Functional Structural Defects

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The authors numerically investigate the optical transmission properties of cholesteric liquid crystals with a local modulation of pitch and refractive indices. The transmittance spectra of structures with various defect thickness, refractive indices and pitch are calculated using the 4×4 matrix method. Calculation results showed that single or multiple transmittance peaks arise in the selective reflection band for light incidence with the same circular handedness as the material, and their spectral positions depend strongly on the physical parameter of the defect. A pitch shortening or elongation at the defect causes the defect mode to shift in the shorter or longer wavelength region, respectively, and the tuning range of the defect mode becomes larger for a thicker defect. We also show that efficient tuning is achieved by a defect medium with a smaller birefringence, allowing the defect mode to be sweeped through the photonic band gap with a smaller modulation of pitch.

Keywords: cholesteric liquid crystals; defect mode; photonic crystals

INTRODUCTION

Photonic Crystals (PhCs) are periodic dielectric media in which light interacts strongly with due to the comparative size of the periodicity of the medium and the wavelength of light. Wavelength regions in which light transmission is inhibited called photonic band-gaps (PBGs) are exhibited in such materials, depending on the refractive index and the periodicity of the comprising material [1,2]. Recently, organic PhCs have been attracting considerable interest due to the

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high potential and ease of fabrication of such devices, unlike inorganic PhCs which generally require a time-consuming and costly process. Liquid crystals (LCs) are particularly interesting materials to be used in PhCs since they are highly birefringent and can easily be reoriented by external stimuli [3], meaning that their physical parameters can be tuned. Numerous theoretical and experimental demonstrations of tunable PhCs using LCs have been made in opal or inverse-opal structures [4,5] or holographic polymer dispersed liquid crystals [6,7]. Introducing LCs in a structural defect of PhCs lead to the realization of a defect mode [8] that can be tuned by external fields: defect-mode tuning [9] and laser action using the cavity effect [10] have been demonstrated for the one-dimensional case, where a LC was infiltrated between two dielectric multilayers.

Cholesteric liquid crystals (ChLCs) in which the liquid crystalline material itself possess a helical periodic structure thus becomes a promising candidate material as a tunable PhC. The fabrication process is extremely simple because the periodic structure is formed spontaneously by the constituent molecules, and owing to the twisted structure of highly birefringent materials, the PBG of ChLCs, or more commonly known as the selective reflection band [11], is larger in ChLCs than in dielectric multilayers with the same refractive indices and pitch (the selective reflection band appears in the wavelength region described by $\lambda = n_o p \sim n_e p$, where n_o and n_e are the ordinary and extraordinary refractive indices, and p is the pitch, while in dielectric multilayers the PBG is narrower depending on the composition ratio of the two materials). Tunability in ChLCs is realized by utilizing the external field sensitivity of the LC molecules or by doping certain functional molecules to induce a change in the refractive indices or the pitch. Thermal and electrical control of the ChLC pitch [12,13] have been demonstrated, and in ChLCs with azobenzene dyes, optical tuning is realized as a result of the trans-cis isomerization of the dye [14].

Both theoretical and experimental investigations have been made on the photonic effects of introducing defects in ChLCs: from introducing a simple isotropic defect layer [15] to an anisotropic nematic LC defect [16] or a phase shift in ChLC directors [17–19]. The conventional method of defect fabrication in ChLCs had been the stacking method where the photopolymerized ChLC and the defect material are stacked on top of each other. We have previously reported a different approach to fabricate defect containing ChLC structures, which was to perform two-photon induced laser lithography in a photopolymerizable ChLC material [20]. This method is advantageous over the stacking method since the size of the defect is limited only by the resolution of two-photon polymerization process which goes down to a few 100 nms (smaller than the stacking process), therefore making it easier to fabricate devices with a narrow defect so that only a single defect mode is exhibited. A ChLC heterostructure fabricated by this method, with an unpolymerized ChLC placed between two polymerized ChLC layers, exhibited a single defect mode with a tunability of over 30 nm upon heating [21]. Another advantage of the two-photon induced laser lithography method is the ease of introducing different materials in the defects. The structure can be template by rinsing out the unpolymerized ChLC and infiltrating another material. We have so far reported an all-optical tunable single mode filter based on azobenzene containing ChLCs introduced as the defect [22].

In the present research, we focus on ChLC heterostructures in which a helical periodic material is introduced as the defect, and theoretically analyze the optical properties upon varying the pitch, refractive indices and the width of the defect layer. We assume that the ChLC pitch and the refractive indices change discretely at the defect, which makes the model different from those presented in previous studies, where the modulation in the refractive indices is neglected, or a spatial pitch modulation with a Gaussian [23] or exponential [24] profile is assumed. The 4×4 matrix method established by Berreman [25] is employed for the numerical calculations in the research.

MODEL AND METHOD OF ANALYSIS

A schematic of the ChLC heterostructure considered in this research is shown in Fig. 1 (a). Within the left-handed ChLC with length d,

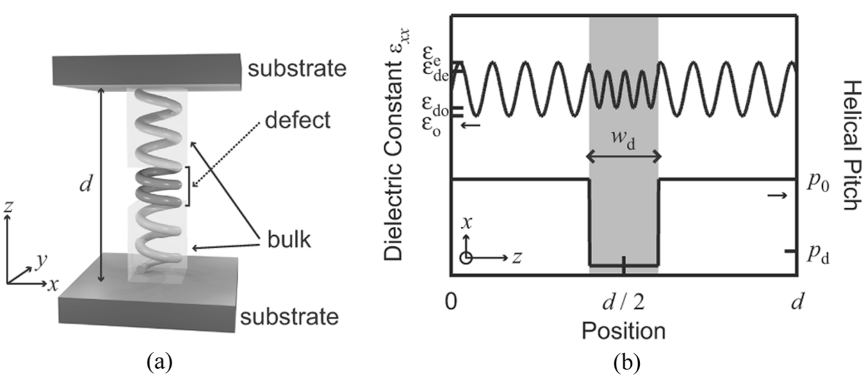

FIGURE 1 Schematic of the director distribution of the ChLC heterostructure (a) and an example of the distribution of the dielectric constant and pitch length (b).

extraordinary refractive indix n_e, ordinary refractive index n_o and pitch length p_0 (bulk ChLC), a different ChLC material with the same handedness as the bulk ChLC but with different refractive indices n_{de} and n_{do}, and pitch p_d (defect ChLC) is confined in a finite region with thickness w_d at the center of the medium ($z = d/2$). A discrete modulation of the pitch and the refractive indices are assumed, following our previous experimental reports [22]. Figure 1 (b) shows an example of the distribution of the pitch and the dielectric constant ε_{xx} of the of the ChLC heterostructure. In the given coordinate system in which the helical axis lies along the z-direction, the distribution of the dielectric tensor ε is given by

$$\mu(z) = \begin{bmatrix} \varepsilon_{xx} & \varepsilon_{xy} & \varepsilon_{xz} \\ \varepsilon_{yx} & \varepsilon_{yy} & \varepsilon_{yz} \\ \varepsilon_{zx} & \varepsilon_{zy} & \varepsilon_{zz} \end{bmatrix} = \begin{bmatrix} \varepsilon_y \sin^2\phi(z) + \varepsilon_x \cos^2\phi(z) & \varepsilon_\alpha \cos\phi(z)\sin\phi(z) & 0 \\ \varepsilon_\alpha \cos\phi(z)\sin\phi(z) & \varepsilon_y \cos^2\phi(z) + \varepsilon_\alpha \sin^2\phi(z) & 0 \\ 0 & 0 & \varepsilon_z \end{bmatrix} \tag{1}$$

where $\varepsilon_x = \varepsilon_e = n_e^2$, $\varepsilon_y = \varepsilon_z = \varepsilon_o = n_o^2$, $\mathrm{d}\phi/\mathrm{dz} = 2\pi/p_0$ and $\varepsilon_\alpha = \varepsilon_x - \varepsilon_y = n_e^2 - n_o^2$ in the bulk region ($|z - d/2| > w_d$) and $\varepsilon_x = \varepsilon_{de} = n_{de}^2, \varepsilon_y = \varepsilon_z = \varepsilon_{do} = n_{do}^2$, $\mathrm{d}\phi/\mathrm{dz} = 2\pi/p_d$ and $\varepsilon_a = \varepsilon_x - \varepsilon_y = n_{de}^2 - n_{do}^2$ in the defect region ($|z - d/2| \leq w_d$). According to Berreman's 4×4 matrix method, light propagating along the z-axis with frequency ω is expressed by a differential equation

$$\frac{d\Psi(z)}{dz} = \frac{i\omega}{c}\mathbf{D}(z)\Psi(z) \tag{2}$$

where $\Psi(z) = [E_x,\ H_y,\ E_y,\ H_x]^T$ and $\mathbf{D}$(z) is the derivative transfer matrix, which is a function of both the dielectric tensor and the wave vector of incident light [25]. The solution to Equation (2) is given by $\Psi(z) = \exp\,[i\omega\mathbf{D}(z)/c]\ \Psi(0)$ and is expanded by a Taylor series to the third power of $\mathbf{D}$(z).

The physical parameters of the bulk ChLC used in this study were $n_e = 1.72$, $n_o = 1.53$ and $P_0 = 380$ nm and were kept constant throughout the course of the simulation. The physical parameters of the defect ChLC were changed so that positive and negative contrast of pitch were introduced ($p_d - p_0 > 0$ and $p_d - p_0 < 0$ respectively) for different refractive index configurations. The total length of the ChLC heterostructure was $d = 5.7\,\mu$m.

DEFECT THICKNESS DEPENDENCE OF THE DEFECT MODE IN DISCRETELY PITCH MODULATED CHLC HETEROSTRUCTURE

We first investigate the effect of the defect thickness on the defect mode wavelength. Here the defect ChLC is assumed to have the same refractive indices as the bulk ChLC for simplicity: $n_{\mathrm{de}} = n_{\mathrm{e}} = 1.72$ and $n_{\mathrm{do}} = n_{\mathrm{o}} = 1.53$. Figure 2 shows the transmittance spectra of the ChLC heterostructure with three different defect thicknesses $w_{\mathrm{d}} = 380\,\mathrm{nm}$, $760\,\mathrm{nm}$ and $1140\,\mathrm{nm}$ and two different defect pitch lengths, $p_{\mathrm{d}} = 330\,\mathrm{nm}$ and $430\,\mathrm{nm}$. One can see a defect mode emerging within the selective reflection band of the bulk ChLC for all defect thicknesse s and pitch lengths, but only for light with the same circular handedness as the ChLC material, which in this case is the left

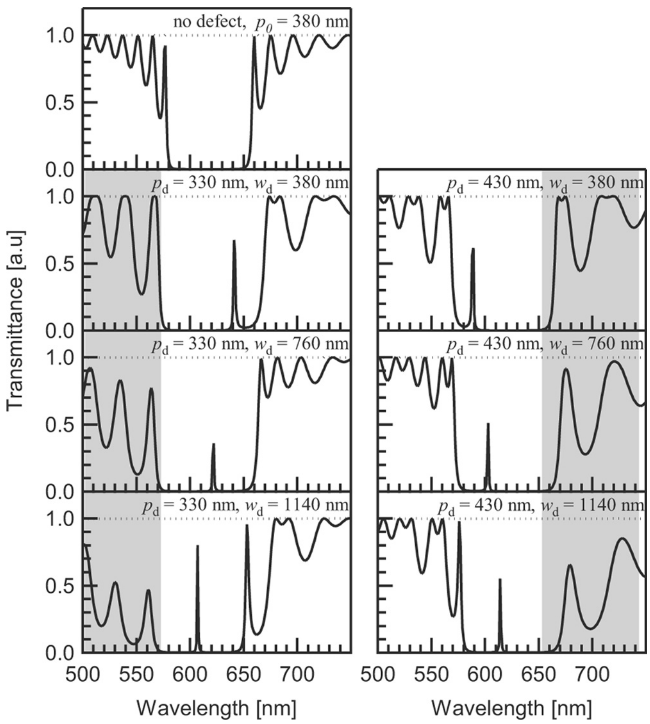

FIGURE 2 Transmittance spectra upon left circularly polarized (solid) and right circularly polarized light incidence, for ChLC heterostructures with defect layers with various thicknesses. Defect ChLCs with both shorter (330 nm) and longer (430 nm) pitch lengths compared to the bulk ($p_0 = 380\,\mathrm{nm}$) are considered but the refractive index is constant throughout the medium. The shaded region indicates the PBG of the defect ChLC.

circularly polarized light (shown in solid lines on graph; dotted line indicates right circularly polarized light incidence). This observation agrees with that obtained experimentally with a helical structure in the defect [21], unlike in the case where isotropic or uniaxial materials are introduced as a defect, affecting both circular polarizations.

The defect modes are generally seen to appear at a "deeper" position in the PBG in structures with a thicker defect layer: that is, they appear at shorter wavelengths than it does in structures with a thinner defect for the case when $p_d < p_0$, while for defects such that $p_d > p_0$, the defect modes appear at longer wavelengths. This is understood in terms of the coupling of light between the two ChLC materials: since the defect mode spatially localizes at the defect and forms a standing wave, they couple to the bandedge mode of the defect ChLC, where standing forms are intrinsically formed [26]. However, since the PBG of the defect ChLC appears at a wavelength shorter (longer) than the PBG of the bulk ChLC for $p_d < p_0$ ($p_d > p_0$) (shaded region in Fig. 2), the defect mode wavelengths shift towards the shorter (longer) wavelength region, closer to the bandedge wavelength, as the coupling becomes stronger upon increasing the thickness of the defect. The effect of the PBG of the defect ChLC also becomes more obvious in thicker defect layers, as seen by the decrease in the transmittance at the wavelengths corresponding to the PBG. One may draw the conclusion that a thicker defect allows a larger tuning range for the same amount of pitch modulation: however, the width of the defect should be decided carefully depending on what kind of characteristics is required, since a defect too wide causes multiple modes to emerge in the PBG. An example is seen in Fig. 2, where a second defect mode appears near the bandedge wavelengths for both cases of $p_d = 330\,\text{nm}$ and $430\,\text{nm}$, when $w_d = 1140\,\text{nm}$.

REFRACTIVE INDEX DEPENDENCE OF THE DEFECT MODE IN DISCRETELY PITCH MODULATED CHLC HETEROSTRUCTURE

The effects of varying the refractive index of the defect ChLC is investigated by calculating the transmittance spectrum of the ChLC heterostructure with different n_{de} and n_{do} values, for cases when a negative and positive contrast of the pitch is introduced. Two types of modulation are considered: the refractive index is varied while keeping the birefringence $\Delta n = n_e - n_o$ consistent with the bulk ChLC, and in the second case the birefringence is varied while keeping the average refractive index $n_{av} = (n_e + n_o)/2$ constant. For the first model we assume (n_{de}, n_{do}, n_{av}) = (1.66, 1.47, 1.565), (1.69, 1.50, 1.595), (1.75, 1.56, 1.655) and (1.78, 1.59, 1.685) and (n_{de}, n_{do}, Δn) = (1.66, 1.59, 0.07), (1.69, 1.56, 0.13), (1.75, 1.50, 0.25) and (1.78, 1.47, 0.31) for the second model. The thickness of the defect is $w_d = 760$ nm and the transmittance spectrum is calculated for different values of p_d from 200–760 nm.

Figures 3 and 4 show the wavelengths of the defect mode for left circularly polarized light incidence as the defect ChLC pitch is modulated, for cases when the birefringence or the average refractive index of the defect ChLC is varied. The unshaded region indicates the PBG of the bulk ChLC and the vertical broken line is the point at which $p_d = p_0 = 380$ nm, and no defect mode exists. Calculation

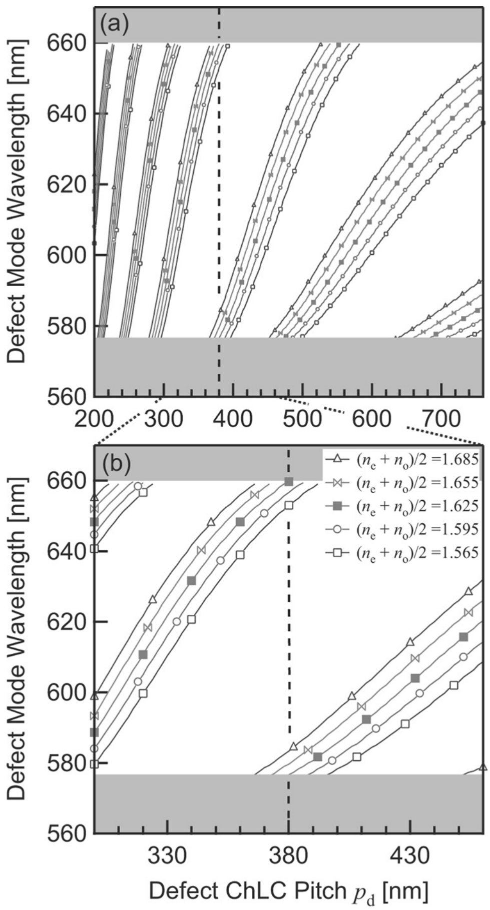

FIGURE 3 Defect mode wavelengths obtained for various values of the defect ChLC pitch p_d, in ChLC heterostructures with a varying average refractive index n_{av}.

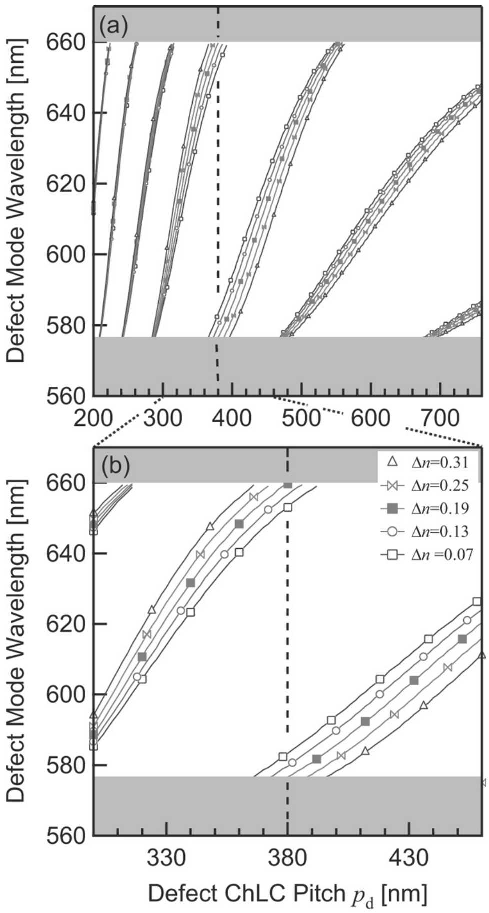

FIGURE 4 Defect mode wavelengths obtained for various values of the defect ChLC pitch p_{d}, in ChLC heterostructures with a varying birefringence Δn.

results for the case when $(n_{\mathrm{de}}, n_{\mathrm{do}}) = (1.72, 1.53)$ (corresponding to the case when the refractive index remains constant throughout the material) is shown for comparison, by filled squares. For either kind of refractive index modulation, blue-shift of the defect mode is observed upon pitch-shortening of the defect ChLC, while a red-shift is observed upon elongating the defect ChLC pitch. This observation originates from the defect modes being coupled to the PBG bandedge mode of the defect ChLC: a shorter (longer) defect ChLC pitch corresponds to a PBG appearing at a shorter (longer) wavelength region, therefore decreasing (increasing) the coupling wavelength.

A comparison of the two figures at different values of p_d reveal several features of the how the refractive index of the defect affects the optical properties of the ChLC heterostructure. In the short p_d limit, one observes that the defect mode wavelength converges for the various refractive index configurations. Since the spatial wavelength of light in the PBG of the bulk ChLC is comparable to its pitch p_0, light does not effectively experience the refractive index modulating with the short periodicity p_d, and the contribution of the average value n_{av} of the refractive index becomes larger than that of the birefringence Δn. This is supported also by the fact that the distribution in the defect mode wavelength is smaller when Δn is modulated (Fig. 4 (a)), compared to the case when n_{av} i s modulated (Fig. 3 (a)). The same phenomena is also observed in the long p_d limit, where the propagating light does not necessarily experience the helical periodicity of the defect ChLC, and the average refractive index contributes largely to the defect mode wavelength.

Values of p_d close to p_0 is of greatest interest in terms of the tuning characteristics, since single-mode tuning can be realized in this region. Figures 3(b) and 4(b) are magnified images of the defect tuning properties for $300 < p_d < 460$ nm. The refractive index of the defect ChLC seems to have a rather large effect on the defect mode wavelength in this region. For either an increase or decrease in the n_{av} of the defect ChLC, the defect mode wavelength is exhibited at a longer or shorter wavelength, respectively, than in the case where no modulation of the refractive index is introduced. For all calculated cases, full range tuning through the PBG of the bulk ChLC can be obtained, although the change in p_d required to tune through the whole PBG is becomes slightly larger for defect ChLCs with a lower n_{av}. For example, full range tuning is obtained in defect ChLCs with $(n_{de}, n_{do}) = (1.78, 1.59)$ by changing the p_d between 306 and 418 nm (112 nm), while for $(n_{de}, n_{do}) = (1.66, 1.47)$ p_d must be swept from 324 to 452 nm (128 nm). For changes in the Δn of the defect ChLC material, different responses are obtained depending on the value of p_d: for $p_d < p_0$, the defect mode appears at a shorter wavelength in defect ChLCs with a larger Δn, while the opposite phenomena is observed for $p_d > p_0$. This means that defect ChLCs with a smaller Δn allows a more efficient tuning; full range tuning is obtained for values of p_d between 316 and 412 nm (98 nm) for $\Delta n = 0.07$, whereas for $\Delta n = 0.31$, p_d must be swept between 312 and 458 nm (146 nm). It should be noted, however, that the tuning range is limited by the birefringence of the defect ChLC material. For $\Delta n = 0.07$, two defect modes appear between $p_d = 366$ and 392 nm, so the effective tuning range for single mode becomes $589\,\text{nm} < \lambda < 648\,\text{nm}$, whereas for large Δn materials complete tuning within the full PBG of the bulk ChLC, that is, between 576 nm and 659 nm is possible. Considering these results, the refractive indices of the defect ChLC should be selected carefully so that the requirements of the optical properties can be met.

It is of interest to compare the tuning characteristics of the defect mode discussed here with another defect characteristic of the helical structure of ChLCs: the twist defect [19]. In the twist defect where there is a discontinuity in the ChLC helix, a single defect mode is exhibited at a wavelength determined by the phase jump. Although full range tuning through the PBG is also possible in these systems by varying the angular twist, the ChLC heterostructure seems to be a more realistic option to realize tunable filters, since the twist defect is introduced by stacking two polymerized ChLCs and requires mechanical tuning, whereas other means of tuning (such as heat [21] or light [22] as mentioned earlier) are possible in ChLC heterostructures.

CONCLUSION

We have numerically described the optical properties of a single-handed ChLC heterostructure in which a defect with a discretely different pitch and refractive indices is introduced. Simulation results indicated that the defect modes are only exhibited for incident light with the same circular polarization as the handedness of the ChLC, and that the defect thickness, refractive indices and pitch all contribute to the defect mode wavelength. Introduction of a defect with a shorter pitch than the bulk caused the defect mode to appear from the longer bandedge wavelength of the PBG of the bulk ChLC and blue-shift as the contrast in the pitch became larger, while the opposite phenomenon occurred for defect ChLCs with a longer pitch. The defect mode appeared at longer or shorter wavelengths for increasing or decreasing the average refractive index of the defect material, whereas more efficient tuning could be achieved by using defect ChLCs with a small birefringence. Our findings provide a strategy which one can follow when designing tunable single mode filters based on ChLC heterostructures with a pitch varying ChLC functioning as the defect.

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