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Incident angle and polarization effects on the dye-doped cholesteric liquid crystal laser

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Abstract

Polarization effect of the pulsed pumping laser on the lasing characteristics of the dye-doped right-handed cholesteric liquid crystal (CLC) is studied at different incident angles. At a small incident angle (<18°), the CLC laser performance is independent of the polarization state of the pumping laser. As the incident angle exceeds 18°, the CLC laser emission strongly depends on the polarization state of the pumping laser. As the incident angle increases, the lasing efficiency of the CLC laser is dramatically decreased for those excited by the right-handed circularly polarized or linearly polarized laser, but barely affected by the left-handed circularly polarized laser. As the incident angle exceeds ~31°, lasing phenomenon diminishes under the excitation of the right-handed circularly polarized laser. The lasing threshold pumped by the linearly polarized laser is ~2× higher than that pumped by the left-handed circularly polarized laser.

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Keywords: Photonic crystal laser; Photonic bandgap; Cholesteric liquid crystal

1. Introduction

Cholesteric liquid crystals (CLCs) have found useful applications for reflective displays, diffusive reflectors, and reflective polarizers [1–3], because of their unique properties, such as supramolecular helicoidal periodic structure and 100% selective reflection of circularly polarized light, and so on. A CLC cell is typically prepared by doping some chiral agents into a nematic liquid crystal mixture. Such cholesteric structure is periodic around a preferred axis and the spatial period is equal to half of the pitch length P_o. Since liquid crystals are highly birefringent media, the periodic helical structure gives a periodic modulation of the refractive index. As a result, a one-dimensional photonic stop band centered at $\lambda_o = nP_o$ is formed. For the incident light falling within the photonic stop band range, only the light with the same circular polarization as the cholesteric helix is strongly reflected; the opposite component is transmitted without any significant reflection.

Based on the photonic band-gap property, a CLC doped with laser dyes can also be used as a mirrorless feedback laser [4–9]. The effects of electric field [4], temperature [5], and light irradiation [6] on the dye-doped CLC lasing performance have been studied. However, the polarization effect of the pump laser on the lasing properties of the dye-doped CLC has not been reported.

In this paper, we investigate the polarization effect of the pump laser on the laser emission of the dye-doped right-handed CLC at different incident angles. When the incident angle is below 18°, the laser emission of the CLC is almost the same no matter the pumping laser is linearly polarized (LP), left circularly polarized (LCP) or right circularly polarized (RCP). As the incident angle exceeds 18°, however, the CLC laser emission strongly depends on the polarization state of the pumping laser. As the incident

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angle further increases, the lasing efficiency dramatically decreases if the pump laser is RCP or LP, but only slightly decreases if the pump laser is LCP. When the incident angle is above 31°, no laser is observed under the excitation of the RCP, and the lasing threshold excited by a LP laser is about twice as that by a LCP laser. The mechanism will be discussed in detail.

2. Sample preparation and experimental setup

The CLC host was prepared by mixing a nematic LC (Merck BL006) with 34 wt% right-handed chiral dopant (Merck CB15) and ∼2 wt% of the fluorescent dye, 4-(dicyanomethylene)-2-methyl-6-(4-dimethylaminostryl)-4H-pyran (DCM, Exciton). The mixture was stirred in isotropic phase for ∼4 h to make the constituents uniformly mixed and then capillary filled in the isotropic phase into a 15-μm thick LC cell. Inside the cell, both the glass substrates were coated with a thin polyimide alignment layer. The anti-parallel rubbing-induced pretilt angle is ∼3°. After the temperature was gradually cooled down to room temperature, a CLC sample with right-hand helix was formed. The pitch length is $P_o \sim 380$ nm and stop bandgap ∼80 nm centered at $\lambda_o \sim 600$ nm.

Fig. 1 shows the experimental setup. A second harmonic Q-switched Nd:YAG pulsed laser (Continuum, Minilite II) at $\lambda = 532$ nm with vertical linear polarization was used to excite the dye-doped CLC sample. The pulse width and repetition rate were 6 ns and 1 Hz, respectively. The reason that we chose 1 Hz repetition rate was to avoid sample heating and degradation. A beam splitter was used to divide the incoming laser beam into two beams. One was detected by a laser energy meter (Ophir, Laserstar) for monitoring the pumping pulse energy and the other was used as the excitation beam. A linear polarizer and a quarter-wave plate were used to convert the linear polarization into right or left circular polarization. A lens with 150 mm focal length focused the incident beam to a small spot size with ∼160 μm diameter at the sample. The output laser emission in the forward direction of the sample was collected by a lens to a fiber-optics-based universal serial bus (USB) spectrometer (resolution = 0.04 nm; USB HR2000, Ocean Optics). Since the output laser emission from the sample is always normal to the sample surface, in order to avoid the optics adjustment during the change of the incident angle, we mounted the sample, lens, head of the fiber-based spectrometer and other necessary optical components including color filters and attenuators on a rail. In this case, the incident angle could be easily changed by rotating the rail without any need of optics adjustment.

3. Experimental results

Fig. 2 shows the transmittance (using an unpolarized light) and lasing emission spectra of the dye-doped CLCs at normal incidence and room temperature. This sample exhibits a reflection band from 550 to 635 nm. The interference fringes beyond the reflection band result from the multiple reflections of the cell. A single lasing peak is observed at the mode closest to the long wavelength edge of the reflection band when the sample is excited by a linearly polarized or circularly polarized pulsed laser.

Since the CLC sample is right hand, it reflects the RCP light within its reflection band but does not affect the propagation of the LCP light. In this study, we would like to

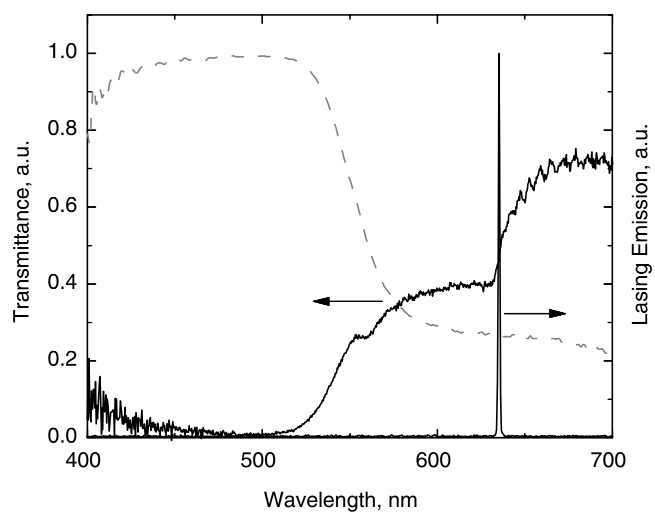

Fig. 2. Transmission spectra of the dye-doped CLC cell and the laser emission line, and the absorption spectrum of the DCM dye (grey dashed line).

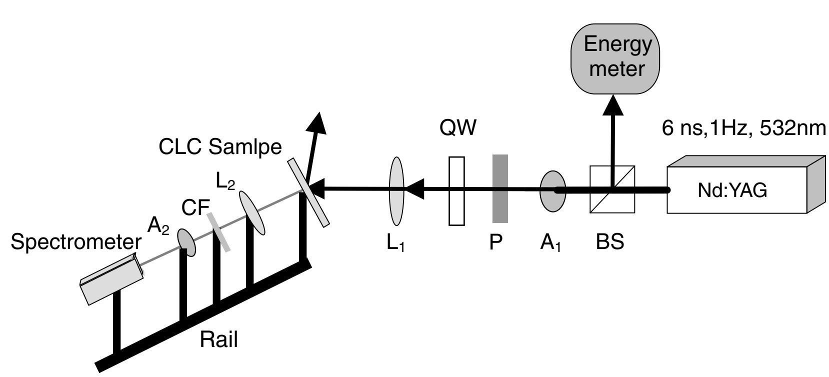

Fig. 1. Scheme of the experimental setup. BS: beam splitter, QW: quarter-wave plate, A: attenuator, CF: color filter, and L: lens.

know how the pump light polarization affects the CLC laser.

Fig. 3(a) depicts the lasing behavior of the CLC sample under the green laser excitation at vertical linear (squares), right (triangles), and left (circles) circular polarization at normal incidence ($\theta = 0°$). As shown in Fig. 3(a), at normal incidence the lasing efficiency is almost the same for all the three different polarization states when compared at the same pumping energy. This indicates that the laser emission is basically polarization independent.

In our experiment, we found that this polarization-independent lasing phenomenon extends to $|\theta| \leqslant 18°$. As the incident angle is above $\pm 18°$, the lasing properties strongly depend on the polarization state of the excitation laser. Here, we take the results at $\theta = 27°$ (in air) as an example to show the polarization dependence. As illustrated in Fig. 3(b), the LCP light has the lowest pumping threshold, followed by the LP light and then the RCP light. Accordingly, at a given green laser energy, the LCP light has the highest efficiency of red laser emission and the RCP light has the lowest efficiency. Further increasing the incident angle by rotating the rail enlarges the difference between the LCP, LP, and RCP pumping lights. Especially when the incident angle is above 31°, laser action stops for the RCP laser pumping. The lasing intensity excited by the LCP light is much higher than that by the LP light, as shown in Fig. 3(c).

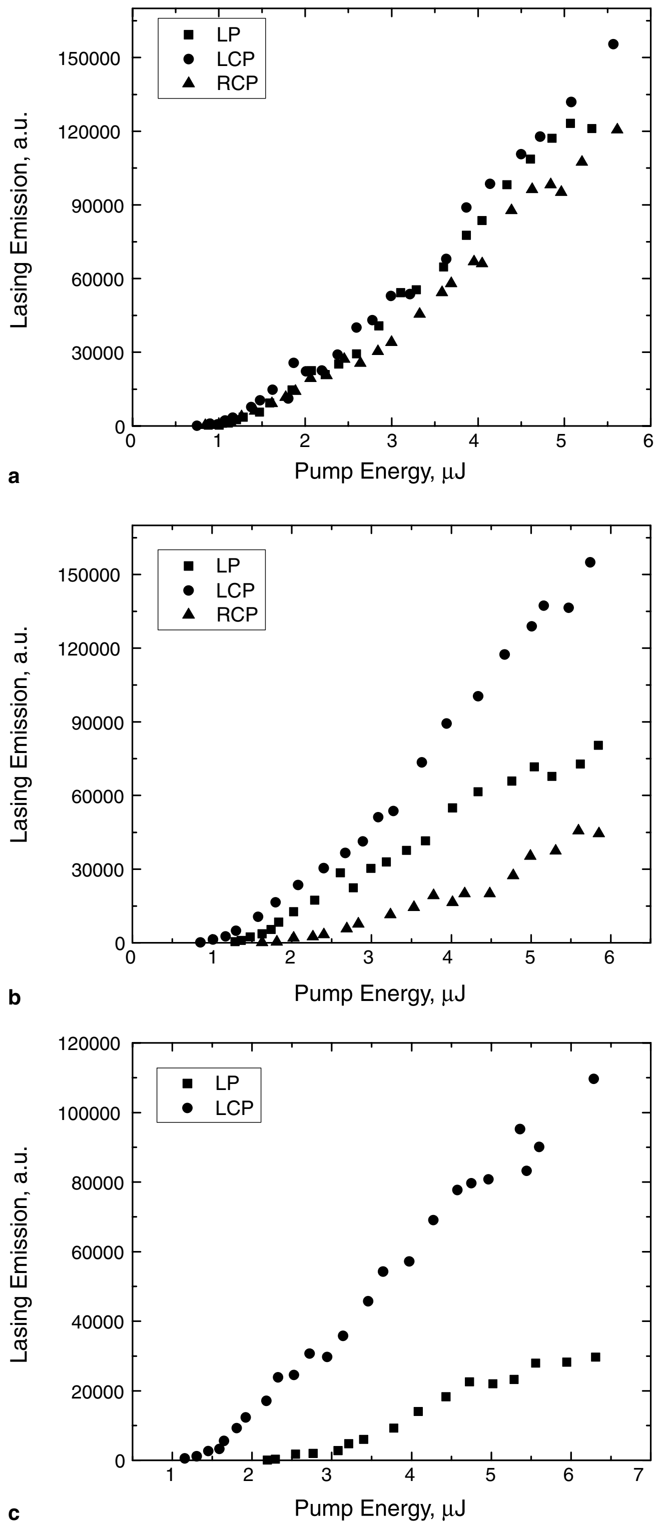

Fig. 3. Laser emission at different incident angles: (a) 0°, (b) 27°, and (c) 31°.

Fig. 4 compares the lasing threshold of the CLC sample excited by the LP, LCP and RCP light at different incident angles. At a small incident angle (<18°), the lasing threshold is insensitive to the polarization of the excitation laser. When the incident angle is above 18°, the RCP (LCP) light has the highest (lowest) lasing threshold. As $\theta > 31°$, no laser can be generated by the RCP pump laser. The lasing threshold of the LP light is almost twice as high as that of the LCP light.

4. Discussion

To understand the above phenomenon, we need to consider the photoexcitation processes of the dye molecules

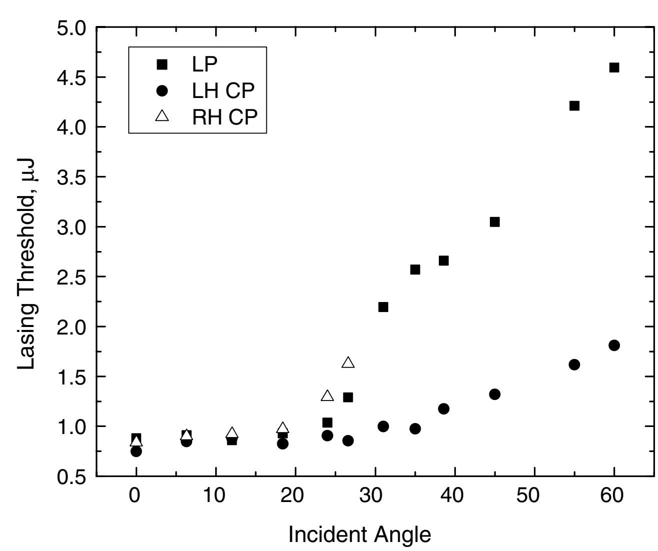

Fig. 4. Lasing threshold excited by the linearly polarized (LP), left-handed (LH), and right-handed (RH) circularly polarized (CP) light at various incident angles.

and the light propagation in the CLC cell at normal and oblique incidences. First, let us examine the photoexcitation process of the dye molecules. Generally, for an isotropic dye molecule, the photoexcitation is insensitive to the polarization of the excitation light. While for an anisotropic dye molecule, the photoexcitation highly depends on the polarization state of the pumping light due to the dichroism of the dye molecules. The photoexcitation of the dye molecules by the linearly polarized pumping light parallel to the dipole moment of the dye molecules is larger than that perpendicular to the dipole moment. If all the dye molecules are oriented in the same direction, the photoexcitation of the sample is polarization dependent. If the dye molecules are randomly distributed in a sample, they function as isotropic media and the photoexcitation is polarization independent. In a CLC sample, the chiral agents possess a twist power. The interaction of the chiral agents and the dipole moment of the LC molecules make the liquid crystal directors twisted to form periodic cholesteric helixes. Accordingly, the mixed dye molecules are also rotated following the cholesteric structure, because of the dipole interactions. As a result, the dye molecules are randomly distributed in the CLC sample, as shown in Fig. 5(a). The dye-doped CLC sample behaves like an isotropic media for the photoexcitation, which can be seen from the transmission spectra of the dye-doped CLC cell for the LCP, RCP, and unpolarized light, as shown in Fig. 5(b). From Fig. 5(b), the dye-doped CLC cell displays different transmission spectra within the 535–750 nm wavelength range for the LCP, RCP, and unpolarized light, because of the existence of the reflection band for the RCP light. While for $\lambda < 535$ nm, these transmission spectra are indistinguishable indicating that the dye itself in the CLC cell exhibits an isotropic absorption spectrum in spite of the effect of the reflection band. Thus, the photoexcitation of the dye-doped CLC sample is polarization independent; so is the lasing emission.

In addition to the photoexcitation of dye molecules, we also need to consider the light propagation in the CLC sample. In a CLC cell, two main factors contribute to the light loss. They are surface reflection and reflection band. First of all, when a light transmits from media 1 with refractive index n_1 to media 2 with refractive index n_2, a portion of light is lost by the surface reflection, as described by the Fresnel formulae [10]:

$$t_{\parallel} = \frac{2\sin\theta_2\cos\theta_1}{\sin(\theta_1+\theta_2)\cos(\theta_1-\theta_2)}, \tag{1}$$

$$t_{\perp} = \frac{2\sin\theta_2\cos\theta_1}{\sin(\theta_1+\theta_2)}, \tag{2}$$

where $t_{\parallel}$ and $t_{\perp}$ are the transmittance of a LP light parallel and perpendicular to the incident plane, respectively, and θ_1 and θ_2 are the incident and refractive angles, respectively. The Snell's law correlates the θ_1 and θ_2 with refractive indices:

$$n_1\sin\theta_1 = n_2\sin\theta_2. \tag{3}$$

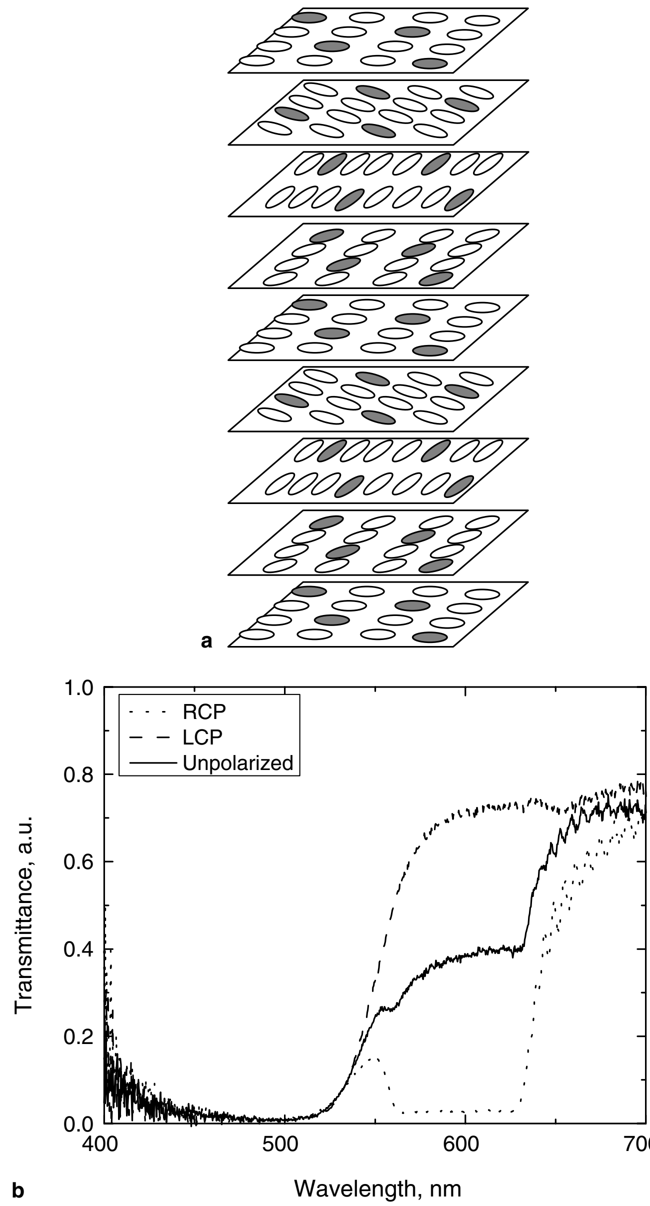

Fig. 5. (a) Distribution of dye (black) molecules in a CLC cell and (b) absorption spectra of the dye-doped CLC for the LCP, RCP, and unpolarized light.

From the above equations, the transmittance does not change too noticeably at small incident angles, but decreases dramatically at a large incident angle [10]. For a CP light, the transmittance (t) can be averaged by

$$t = \frac{(t_{\parallel}+t_{\perp})}{2}. \tag{4}$$

Therefore, the transmittance of the circularly polarized light shows almost the same trend as the linearly polarized light regarding to the incident angles.

As for the light loss induced by the CLC's reflection band, it is known that at the normal incidence the helical pitches of the CLC layers introduce the Bragg reflection center at wavelength λ_o:

$$\lambda_o = np_o. \tag{5}$$

In Eq. (5), p_o is the CLC pitch length and n is the average refractive index defined by

$$n = (n_e + n_o)/2, \tag{6}$$

where n_o and n_e are the LC refractive indices of the ordinary and extraordinary rays, respectively. For an oblique incidence wave propagating at an angle of Θ inside the LC media, the central wavelength of Bragg reflection is governed by

$$\lambda = nP_o \cos\Theta, \tag{7}$$

where Θ can be correlated with incident angle in air θ by Snell's law as

$$\Theta = \sin^{-1}\left(\frac{1}{n}\sin\theta\right). \tag{8}$$

From Eqs. (7) and (8), blue shift occurs as the incident angle tilts away from normal.

Fig. 6(a), (b) and (c) are the plots of the transmission spectra of the LH and RCP, and LP light, respectively, through a CLC cell without dye at various incident angles. First, let us discuss the transmission spectrum of the LCP incident light, as shown in Fig. 6(a). There is no stop band for the LCP light. However, due to the substrate surface reflections, the transmittance reaches only ~0.8 in the visible region at $\theta = 0°$. For the incident angle $\theta \leqslant 18°$, the sample transmittance is only slightly decreased with the increased incident angle. However, for $\theta > 18°$, the sample transmittance dramatically decreases as the incident angle increases. This explains why the lasing threshold induced by the LCP light is barely changed if the incident angle is less than 18°, but is dramatically increased when the incident angle is above 18°.

While for the RCP incident light, there exists a stop band ranging from 550 to 635 nm at normal incidence, which does not cover the excitation laser wavelength at $\lambda = 532$ nm. The light loss is mainly due to the surface reflections, same as LCP light. Since the photoexcitation of the dye-doped CLC sample is polarization independent, the lasing efficiency induced by the RCP light is the same as that by the LCP light at normal incidence, as shown in Fig. 3(a). As the incident angle increases, besides the increased light loss induced by the substrate surface reflections in the whole visible spectral region, the stop band also produces a noticeable blue shift, as shown in Fig. 6(b). When the incident angle is less than 18°, although the stop band produces blue shift, the band gap does not yet cover the excitation wavelength $\lambda = 532$ nm. When the incident angle is above 18°, the short edge of the band gap starts to cover the pumping wavelength $\lambda = 532$ nm, indicating a portion of the RCP light of the pump light is reflected by the sample. Further increasing the incident angle causes more RCP pump light to be reflected, because the center of the stop band moves towards $\lambda = 532$ nm. This explains well our experimental results shown in Fig. 3(b): the lasing efficiency is barely changed when the incident angle increases from 0° to 18°, but is dramatically decreased as the incident angle exceeds 18°. When the incident angle is increased to ~31°, the stop band covers $\lambda = 532$ nm due to the blue shift. As a result, most of the RCP pump light is reflected by the sample. Because of this, few photons in the right circular polarization can enter the sample to excite the dye molecules. Therefore, no lasing can be generated by

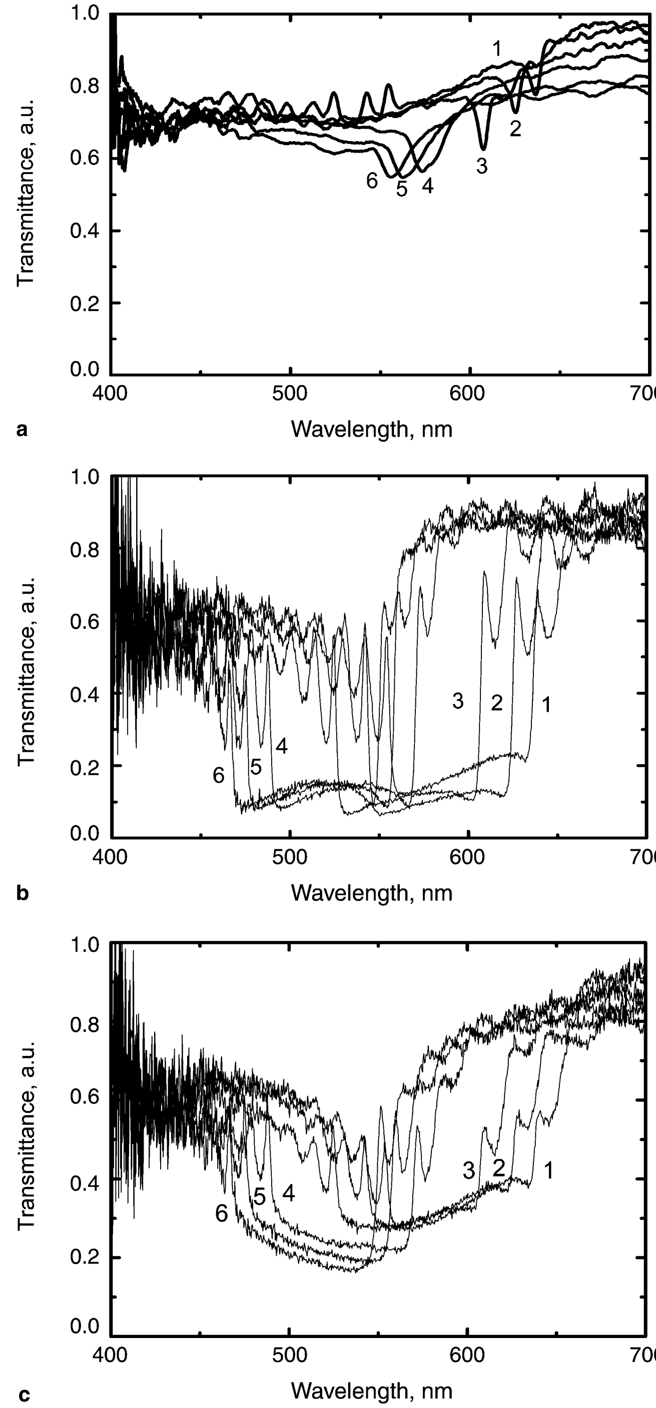

Fig. 6. Transmission spectra of the CLC without dye for (a) left-handed circularly polarized light, (b) right-handed circularly polarized light, and (c) linearly polarized light at different incident angles. Curves 1, 2, 3, 4, 5, and 6 are corresponding to the incident angle at 0°, 18°, 31°, 48°, 54°, and 60°, respectively.

the RCP pump light for the incident angle above 31°, as shown in Fig. 3(c).

A LP light can be decomposed into a right-handed and a left-handed circularly polarized light. Thus, a LP light also displays a stop band which produces blue shift as the incident angle increases, as shown in Fig. 6(c). The laser emission and threshold induced by LP light also shows the same trend to the incident angle as that by the RCP light. It is barely changed by the variation of the incident angle when the incident angle is below 18° and dramatically decreased as the incident angle exceeds 18°. Different from the RCP light, the LP light can still generate lasing output from the sample at incident angle above 31° due to the contribution of the other half LCP light. At a large incident angle (>18°), additional optical loss of the RCP light occurs besides the surface reflections. Thus, the lasing threshold of a LP excitation light is higher than that with LCP pumping light, as seen in Fig. 4.

5. Conclusion

We have investigated the pump laser polarization effect on the lasing performance of the dye-doped CLCs at various incident angles. At small incident angles (<18°), the lasing efficiency of the CLCs is insensitive to the pumping light polarization, and is inert to the variation of the incident angles. At larger incident angles (>18°), however, the lasing performance of the CLC laser strongly depends on the polarization state of the pulsed pump laser. With the increase of the incident angle the lasing efficiency is dramatically decreased if the dye-doped CLC is pumped by a RH circularly or a linearly polarized pulsed laser, but only decreased slightly if it is excited by a LCP laser. As the incident angle exceeds 31°, lasing does not occur under the excitation of a RCP pulsed pump laser. As incident angle continues to increase, the lasing efficiency is dramatically decreased for the LP and the LCP pump laser due to the light loss induced by the blue shift of the reflection band and surface reflections. Therefore, the lasing threshold by a LP light or a LCP excitation laser increases with the increased incident angle. Moreover, the lasing threshold by a LP pump laser is about twice as that by a LCP pump laser. The results give us valuable insights on how to avoid unnecessary light loss in order to optimize the CLC lasers. It would be better to use a laser beam with the opposite circular polarization to the CLC helix to excite the sample at small incident angle.

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