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Fabrication of multi-pitched photonic structure in cholesteric liquid crystals based on a polymer template with helical structure

Jinbao Guo,[a] Hao Wu,[a] Fengjin Chen,[a] Lipei Zhang,[b] Wanli He,[b] Huai Yang[b] and Jie Wei*[a]

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In this study, we demonstrate a novel method for fabricating a single-layer Ch-LC film with multi-pitched photonic structure as new photonic band gap (PBG) materials by utilizing a polymer template. The polymer template with helical structure was originated from polymer-stabilized cholesteric liquid crystals. Various types of Ch-LCs are subsequently imbibed into the polymer template, resulting in films with distinct optical characteristics. Simultaneous red, green and blue reflection (multiple PBGs) and wide-band reflection in a single-layer Ch-LC film can be achieved by using nematic liquid crystals with different birefringence in Ch-LCs/polymer template composites, and a H-bonded Ch-LC with a great temperature-dependence of pitch length was introduced into the polymer template, and a temperature-tunable PBG of the Ch-LC film with a double-handed circularly polarized light reflection band was obtained. Additionally, the lasing emission of a single-layer Ch-LC film and patterning of a single-layer Ch-LC film with a double-handed circularly polarized light reflection band were also realized by the above approach. These special optical properties make the novel Ch-LCs composites interesting for their great potential applications in many fields, such as information recording, optical components, flat displays, photonic crystals *etc*.

Introduction

Photonic crystals (PCs) with an ordered periodic structure at optical wavelengths' scale have attracted a lot of interest from both fundamental study and practical application.[1–5] One of the important forms of PCs is cholesteric liquid crystals (Ch-LCs) that spontaneously form periodic helical structures, which lead to selective reflection for circularly polarized light.[6–13] In Ch-LCs, the period of the helicoidal structure is equal to half the pitch p, and for light propagating along the helical axes, $p = \lambda_0 \times n$, where λ_0 is the wavelength of the maximum reflection or the middle of selective reflection band, and n is the average refractive index $n = (n^e + n^o)/2$. The extraordinary and ordinary indices of refraction are denoted by n^e and n^o, respectively. The photonic bandgap (PBG) width of a conventional Ch-LC is equal to $p \times \Delta n$, and is proportional to the anisotropy of refractive indices $\Delta n = n^e - n^o$. Within the bandwidth, right-circularly polarized light is reflected by a right-handed helix, whereas left-circularly polarized light is transmitted. Outside the bandwidth both polarization states are transmitted.[14,15] Tuning the PBG characteristics of Ch-LCs, such as tuning λ_0 in the visible or infrared spectrum, increasing the PBG width, or increasing reflected light flux, is attractive for many applications in reflective displays, polarizers and color filters.[16–33] In addition, lasing from dye-doped Ch-LCs as a novel photonic application has been extensively studied.[6–12]

[a]College of Materials Science and Engineering, Beijing University of Chemical Technology, Beijing 100029, P. R. China. E-mail: weijie-2008@hotmail.com
[b]State Key Laboratory for Advanced Metals and Materials, Department of Materials Physics and Chemistry, School of Materials Science and Engineering, University of Science and Technology Beijing, Beijing 100083, P. R. China

As is well known, the periodic helical structure of a Ch-LC can be controlled with external influences including electrical fields, temperature, optical radiation and so on.[13,16–21] Additionally, introduction of a photonic defect into a PBG structure of a Ch-LC, such as insertion of an isotropic/anisotropic material with different refractive index, can also be used to modulate the PBG characteristics of the Ch-LC system.[34–37] For example, introduction of a polymer-dye-doped nematic layer between two Ch-LC films can result in modulation of reflectance over 50% and efficient lasing emission.[34,35] Recently, a novel multilayered film consisting of a single-pitched Ch-LC and isotropic PVA was fabricated, and simultaneous red, green and blue reflections (multiple PBGs) were demonstrated.[36] Multiple PBGs lasing emissions were further achieved by introducing two fluorescence dyes in the above system.[37] The above sandwich structure, however, has some limitations, such as complicated structure design, control of optical defects and losses at the interfaces.

In this study, we described a novel method for fabricating a single-layer Ch-LC film with multi-pitched photonic structure by utilizing a polymer template and various types of Ch-LCs that exhibit many special optical reflection properties. Herein, the polymer template with helical structure was prepared by photopolymerizing LC monomers that were dispersed within a Ch-LC mixture, which initially defined the periodicity or pitch (P) of the structure, but was then subsequently removed. Because of memory effects attributable to the polymer template with helical structure,[38,39] when various types of Ch-LCs, including Ch-LCs with different handedness and H-Bonded Ch-LCs, are subsequently refilled into the polymer template, multi-band and wide-band reflection, temperature tunable PBGs of Ch-LCs with a double-handed circularly polarized light reflection band, lasing emission of a single-layer Ch-LC film with a double-handed circularly polarized light reflection band, and the patterning of a single-layer Ch-LC film with a double-handed circularly polarized light reflection band can be achieved. The main practical purpose of this work is to develop a facile and effective strategy leading to the fabrication of single-layer Ch-LC films with multi-pitched photonic structures for photonic and optical applications.

Experimental section

Materials

In this study, non-reactive LCs SLC-1717 (20 °C, 589 nm, $\Delta n = 0.201$) and SLC-6830A (20 °C, 589 nm, $\Delta n = 0.09$) (Slichem Liquid Crystal Material Co., Ltd.); nematic diacrylate monomer, C6M, LC242 and LC1057; diacrylate chiral monomer (DCM), whose phase transition temperatures are Cr–Ch, Ch–I, 57 and 89 °C, respectively; H-bonded Ch-LC (HB-CLC), cholesteryl isonicotinate (proton acceptor)/4-hexyloxy-benzoic acid (proton donor), whose phase transition temperatures are Cr–SmA, SmA–TGBA*, TGBA*–Ch, Ch–I, 127, 155, 157 and 195 °C, respectively; chiral dopant, S811/R811 (Merck Co. Ltd) and photoinitiator, 2,2-dimethoxy-2-phenyl-acetophenone (Irgacure 651, TCI Co. Ltd.); lasing dye (LD), 2-{2,6-bis-[2-(4-dimethylamino-phenyl)-vinyl]-pyran-4-ylidene}-malononitrile were used. Nematic diacrylate monomer C6M was synthesized according to the method suggested by D. J. Broer *et al.*,[40] and another two nematic diacrylate monomers, LC242 and LC1057, were purchased from BASF Co. Ltd. DCM and HB-CLC were prepared as described in our early paper.[41,42] LD was provided by the Technical Institute of Physics and Chemistry, Chinese Academy of Sciences of China. Fig. 1 shows the chemical structures of C6M, DCM, HB-CLC, S811/R811, LD and Irgacure 651.

Experimental cells

In order to induce a planar orientation of LC molecules, the inner surfaces of indium tin oxide coated (ITO) glass cells were coated with a 3.0 wt% polyvinyl alcohol (PVA) aqueous solution.

Table 1 Compositions of Ch-LC mixtures and the reflection band of the cells obtained

Mixture (type)	Weight ratio/wt%	λ_M/nm[a] Before	λ_M/nm[a] After	T_{ChI}/°C[b]
LHHS				
1	SLC-1717 (70.0)/DCM (10.0)/S811 (15.0)/C6M (5.0)	770	705	75
2	SLC-6830A (72.0)/DCM (10.0)/ S811 (13.0)/C6M (5.0)	763	702	73
3	SLC-6830A (53.0)/DCM (10.0)/ S811 (32.0)/C6M (5.0)	478	468	64
4	SLC-6830A (72.7)/S811 (27.3)	551	—	62
RHHS				
5	SLC-1717 (62.3)/R811 (22.7)/C6M (15.0)	593	537	67
6	SLC-1717 (66.0)/R811 (19.0)/C6M (15.0)	756	717	71
7	SLC-1717 (69.0)/R811 (16.0)/C6M (15.0)	765	704	77
8	SLC-1717 (83.5)/R811 (16.0)/0.5 (LD)	696	—	70
9	SLC-1717 (67.5)/R811 (32.5)	497	—	60

[a] Reflection band before and after UV irradiation. [b] $T_{Ch\text{-}I}$ clearing temperature from cholesteric to isotropic phase, which is determined by polarizing light microscope.

The deposited film was dried at 80 °C for 30 min and subsequently rubbed with a textile cloth under a pressure of 2.0 g cm^{-2} along one direction. PET (polyethylene terephthalate) films of 25 μm thickness were used as the cell spacers. The samples were filled into the cells by capillary action at an appropriate temperature. The compositions of the samples are listed in Table 1.

Fabrication of the polymer template

The polymer template was prepared by carrying out the following procedure. At first, the cell containing the sample was irradiated with 365 nm UV light for 30 min for polymerization

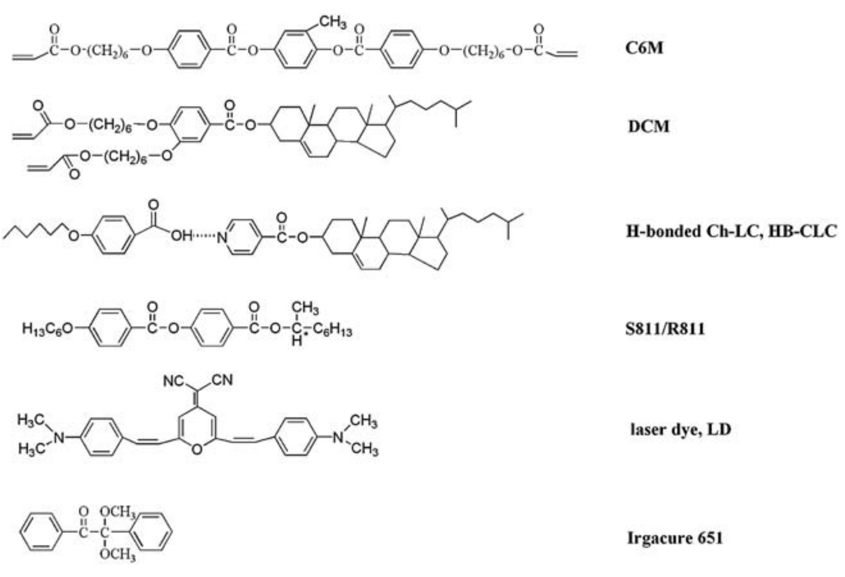

Fig. 1 The chemical structures of the materials used.

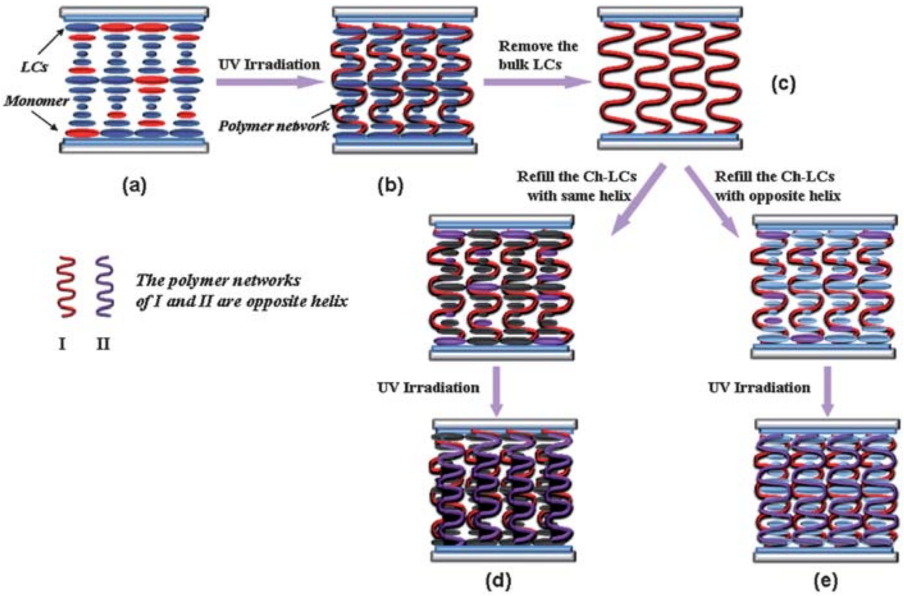

Fig. 2 Illustration outlining the procedure for preparing the Ch-LCs film.

purposes, as shown in Fig. 2(a) and (b), in which the sample was composed of photopolymerizable monomers/non-reactive Ch-LC/photo-initiator with the periodicity or pitch (P) defined by the Ch-LC. Following that, the cell was immersed in cyclohexane for about 48 h, and later in tetrahydrofuran for 20 min to remove the non-reactive LCs. After that, the cell was kept in a vacuum chamber at 60 °C for about 3 h. Thus, the polymer template was obtained as shown in Fig. 2(c). Then, the cell containing the polymer template was refilled with samples with a different kind of Ch-LCs by a capillary filling process and followed by other process, such as second irradiation as shown in Fig. 2(d) and (e).

Lasing of Ch-LC film reflecting both left- and right-circularly polarized light

The pumping source of the dye-doped Ch-LC cells was a single pulse of the second-harmonic generation (SHG) (532 nm) from a Q-switched Nd:yttrium–aluminium–garnet (YAG) laser (Spectra-Physics, Pro-230). The duration of the Q-switched pulse was about 7 ns. The pumping laser was focused on to the sample at an incidence angle of 45° to the normal of the surface, using a lens with a focal length of $f = 10$ cm. The Ch-LC cells were lased in the direction of the surface normal. The emission spectra were recorded with an Ocean Optics HB-2000 spectrometer using fiber optics.

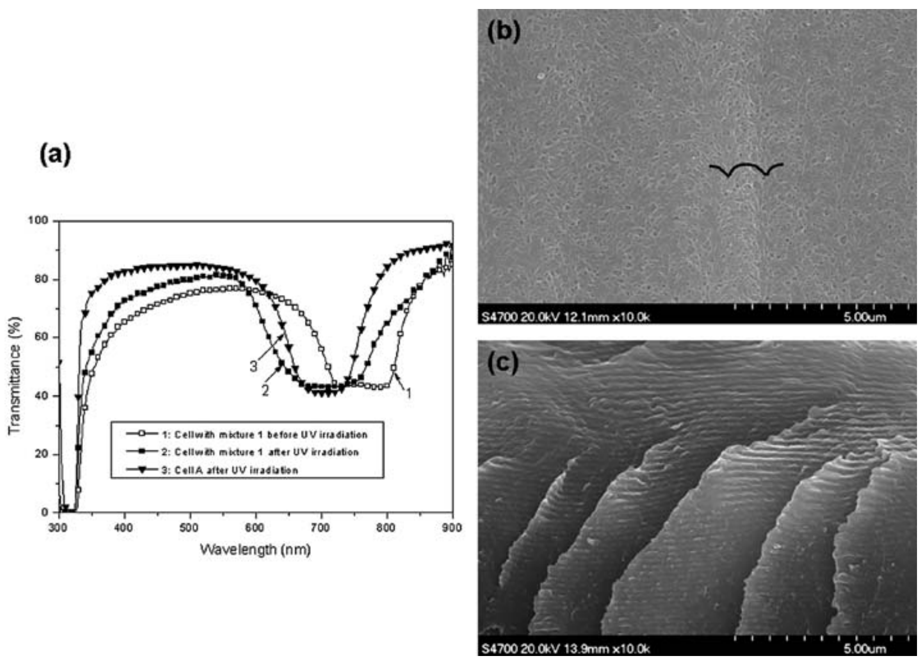

Fig. 3 (a): The transmission spectra of a cell with mixture 1 before and after UV irradiation and cell A after irradiation; (b): SEM micrograph of the surface morphology of the cell with mixture 1 after UV irradiation; (c): SEM micrograph of the cross-section of cell A after irradiation.

Measurements

The samples were observed using a polarizing light microscope (POM) (Olympus, BX51). The optical images were recorded using Linksys 2.43 software. The transmission spectra were obtained by UV/VIS spectrophotometer (Hitachi, U-3010) at normal incidence. The transmittance of a blank cell was normalized to 100%. The minimum wavelength of the transmitted light and the bandwidth at half-height of the peak are defined as T_M and $\Delta\lambda$, respectively. The fluorescence measurements were performed at room temperature using a Hitachi F-4500 spectrometer. The morphology of the polymer network was examined by scanning electron microscopy (SEM) (Hitachi, S-4700). The samples for SEM studies were prepared according to the method described in a previous study.[29]

Results and discussion

Memory effect of the polymer template with helical structure

For our study, a polymer template with helical structure was achieved by photo-polymerizing LC monomers that were dispersed within a Ch-LC mixture, followed by forming polymer-stabilized cholesteric liquid crystals (PSCLCs) which initially defined the periodicity or pitch (P) of the structure, then subsequently removed the non-reactive LCs from the system as displayed in Fig. 2. It should be pointed out that the helix-structure memory of the polymer template which is derived from the initial PSCLCs plays an important role in this study, in which the polymer network is to template the liquid crystalline order by transferring the respective structure and orientation onto the network during polymerization[38,39,43,44] As a representative illustration, a cell with mixture 1 was used to illustrate the above point, Fig. 3(a) shows the transmission spectra of a cell with mixture 1 before irradiation (curve 1) and after irradiation (curve 2), and as shown in Fig. 3(b), the helical superstructure of the polymer network formed in PSCLCs can clearly be imaged from the surface morphology of the cell with mixture 1 after irradiation. Furthermore, cell A was obtained by infiltrating a photo-polymerizable LC mixture into the cell with mixture 1 after irradiation and removal of non-reactive LCs thereafter, in which the photo-polymerizable nematic LC mixture was prepared by mixing C6M, LC242 and LC1057 at a weight ratio of C6M : LC242 : LC1057 = 1 : 1 : 1 and 2% by weight of monomers of photoinitiator Irgacure 651 is added. The transmission spectra of the cell A after UV irradiation at 50 °C is shown in Fig. 3(a), it is obvious that a reflection band reappeared at about 705 nm in the spectra (curve 3) which is approximate to that of the cell with mixture 1 after irradiation (curve 2). Fig. 3(c) shows a SEM image of the cross-section of cell A after irradiation. The distribution of the pitch length of integrated solid cholesteric film can be observed clearly and the pitch length of the Ch-LCs is about 0.45 μm. From the above discussion, we can confirm further that the polymer template has a characteristic of memorizing the periodicity or P of the structure in the initial PSCLCs.

Multi-band reflection and wide-band reflection in a single-layer Ch-LC film

In general, a Ch-LC is a self-assembled PC material that has a single-pitched structure and an associated single-color reflection band for circularly polarized light with the same handedness as the Ch-LC helix. Therefore, simultaneous red, green and blue reflection (or multiple PBGs) in a single-layer Ch-LC film has attracted much attention recently.[36] In this study, the polymer template method was engaged to achieve a simultaneous red, green and blue reflection (multiple PBGs) in a single-layer Ch-LC film with multi-pitched structure. Herein, mixtures 2, 3 and 4, which are all left-handed structures, were used as shown in Table 1, where Δn of the non-reactive LCs in these three mixtures, SLC-6830A, is small and equal to 0.09. In the process of preparing the Ch-LC film with multiple PBGs, the optimum technological parameters are as follows: polymerization temperature is 30 °C, irradiation intensity is 0.65 mW cm^{-2} and exposure time is 15 min. Fig. 4(a) shows the transmission spectra of a cell with mixture 2 before and after UV irradiation, a cell with mixture 3, and cell B. For the cell with mixture 2, its reflection wavelength shifts from 763 nm (curve 1) to 702 nm (curve 2) as a result of the formation of the polymer network in the bulk LCs after UV irradiation, due to the polymerization and crosslinking reaction-induced volume shrinkage.[31,45] The reflection wavelength of the cell with mixture 3 is about 478 nm

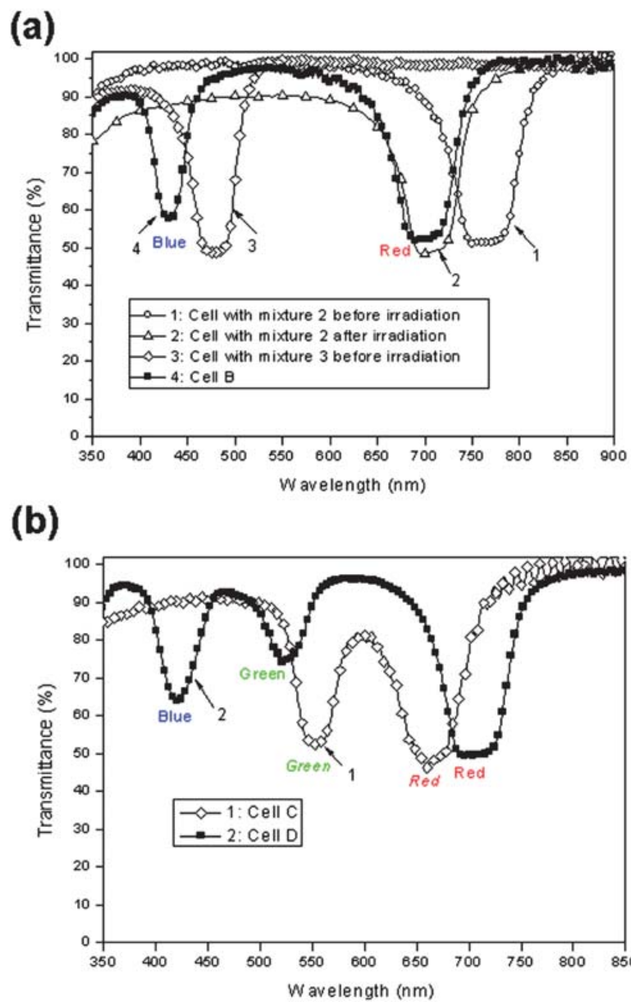

Fig. 4 (a): The transmission spectra of a cell with mixture 2 before and after UV irradiation, a cell with mixture 3 before irradiation, and cell B; (b): the transmission spectra of cell C and D.

(curve 3). After SLC-6830A was washed off and refilled with mixture 3, we were able to observe two reflection bands in the spectra of the cell B (curve 4) occurring due to the difference in the pitch lengths of the polymer network and the bulk LC (mixture 3). That is, after the above procedure, a red/blue colored reflecting Ch-LC film was successfully prepared with the reflection wavelength located at 698/430 nm (curve 4), as shown in Fig. 4(a). Furthermore, cell C was obtained by infiltrating mixture 4 into the cell with mixture 2 after irradiation and extraction of non-reactive LCs thereafter, whereas cell D was achieved by refilling mixture 4 into cell B after irradiation and removal of non-reactive LCs thereafter. As a result, red/green and red/green/blue colored reflecting Ch-LC films were also successfully achieved with the reflection wavelength located at 662/550 nm (curve 1, cell C) and 705/522/423 nm (curve 2, cell D) in Fig. 4(b), respectively. As we can see, the reflection intensity of cell D at the 522 nm reflection band was influenced by the denser polymer network after two-step UV irradiation. However, we can choose the appropriate concentration of the polymer network to improve this condition. Additionally, the location of the reflection band can by adjusted by changing the addition of the chiral dopant. The present system can extend the applications of Ch-LCs to a wide region and could give rise to new photonic devices, in which white or multi-color light is manipulated.

Additionally, increasing birefringence (Δn) of the nematic LCs in the polymer template technique can realize wide-band reflection of the Ch-LC molecules. As is known to all, some Ch-LC materials, which mainly are distinguished as two classes, have been exploited to broaden the PBG bandwidth ($\Delta\lambda$) of Ch-LC molecules. One is the Ch-LC polymer films with a pitch gradient

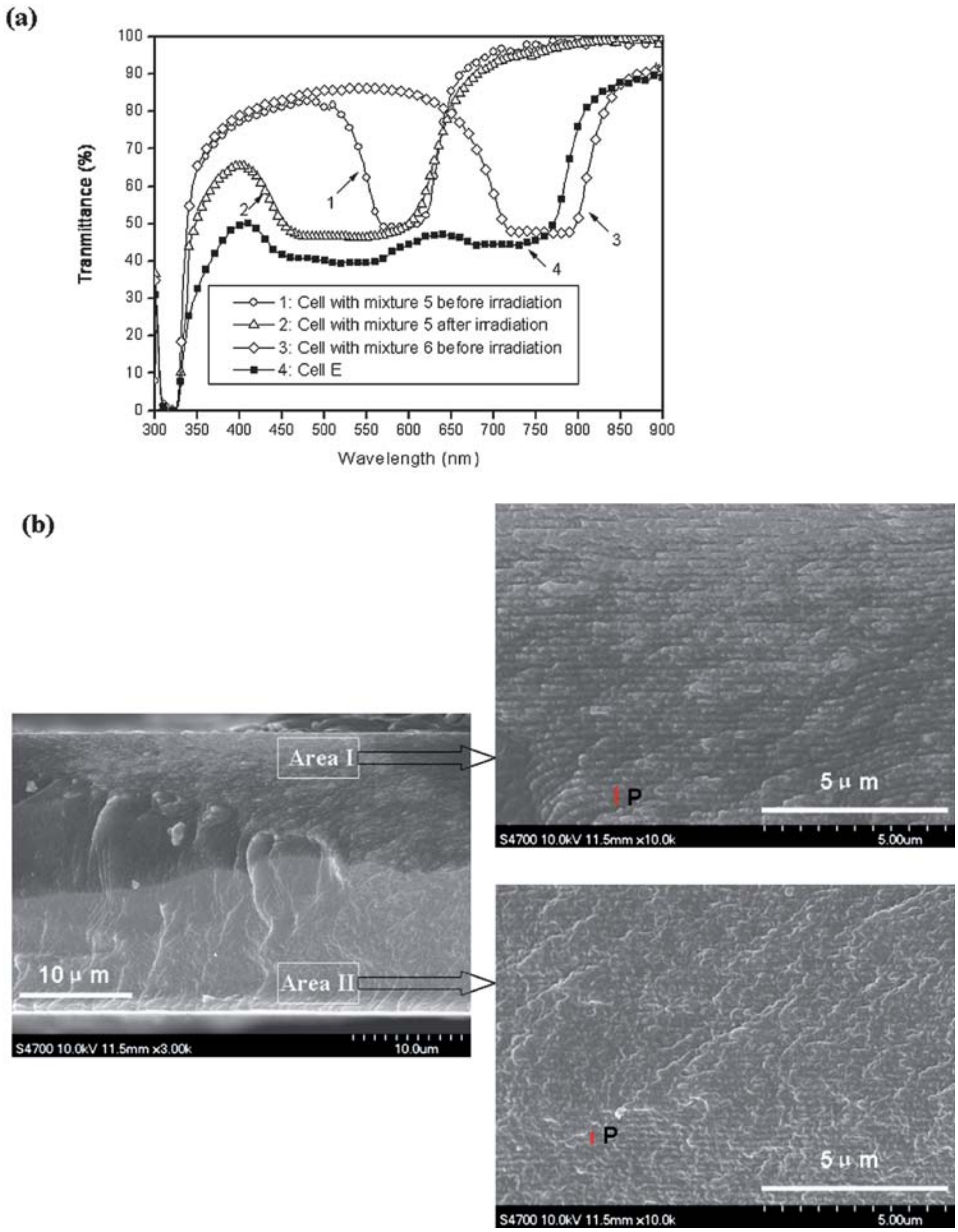

Fig. 5 (a): The transmission spectra of a cell with mixture 5 before and after UV irradiation, a cell with mixture 6, and cell E.; (b): the SEM image of the cross-section of cell F.

or a pitch non-uniform distribution achieved from photo-polymerization of LC acrylates.[22–24] Second is the polymer-stabilized Ch-LC films in which a pitch gradient of a low molar mass Ch-LC can be stabilized by the crosslinked network dispersed within the LC matrix.[25–30] In this study, overlapping of the PBGs of two Ch-LCs materials with different pitch-length and subsequent UV irradiation in the polymer template method was used to obtain wide-band reflection in a single-layer film as shown Fig. 2(d). Here, mixtures 5 and 6, which are both right-handed structures, were used as shown in Table 1, where Δn of non-reactive LCs, SLC-1717 in these two Ch-LC mixtures, is 0.201. When preparing the wide-band reflection Ch-LC films, the technological parameters are as follows: polymerization temperature is 35 °C, irradiation intensity is 0.15 mW cm^{-2} and exposure time is 30 min. Fig. 5(a) shows the transmission spectra of a cell with mixture 5 before and after UV irradiation, a cell with mixture 6, and cell E. For the cell with mixture 5, its reflection band after UV irradiation (curve 2) is much broader than that of the cell before UV irradiation (curve 1), which we believe is due to the natural ultraviolet light absorbing properties of the liquid crystal constituent as suggested by Mitov *et al.*[30] Then, washing out SLC-1717 and infiltrating mixture 6 followed by UV irradiation, it can be seen in Fig. 5 (curve 4) that the bandwidth of the cell E can reach 340 nm from 450 to 790 nm due to the overlapping of two reflection bands of the above Ch-LC mixtures 5 and 6, which includes red, green and blue lights in the visible light spectra. Fig. 5(b) shows the SEM image of the cross-section of cell F, in which cell F was obtained by infiltrating the above photo-polymerizable nematic LCs mixture into cell E after removal of non-reactive LCs, and followed by UV irradiation. As can be seen in Fig. 5(b), the pitch lengths in different regions are different, and the difference between the maximum and the minimum values are approximately equal to those of the Ch-LCs of mixtures 5 and 6, respectively. This demonstrates that the above explanation is reasonable.

Thermally controllable PBGs of Ch-LCs with a double-handed circularly polarized light reflection band

Many efforts have been devoted to adjust the selective reflection (PBGs) of the Ch-LCs by changing external factors including electric, magnetic, and acoustic fields, temperature, and light irradiation as mentioned above.[13,16–21] As is well known, hydrogen bonding, as a non-covalent approach, has been used for the preparation of a wide variety of self-assembled systems because of its stability, dynamics, directionality, and reversibility.[46] In this study, a H-bonded Ch-LC (HB-CLC, as shown in Fig. 1) with a left-handed helix was used to modulate the reflection band of the Ch-LC. Based on its thermo-chromism performance in the visible region, as well as the facileness of preparation, the PBG of a Ch-LCs film with a double-handed circularly polarized light reflection band can be reversibly

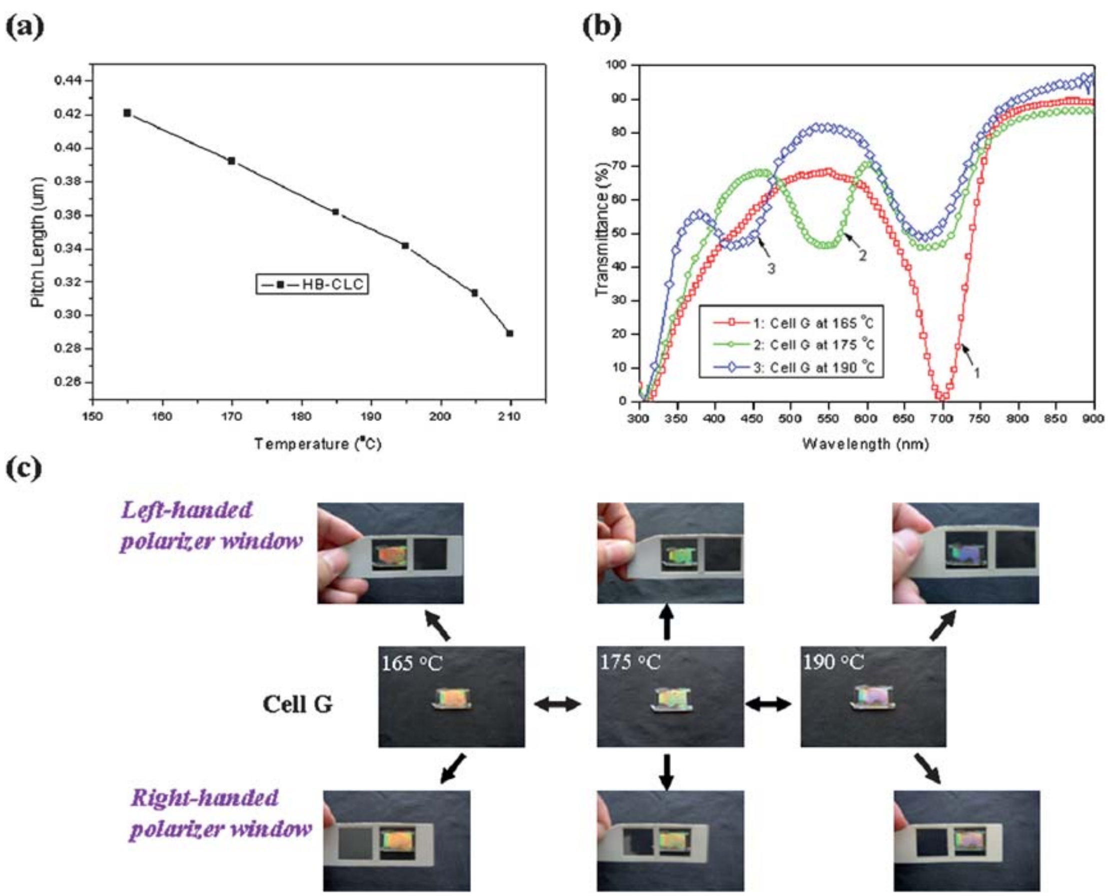

Fig. 6 (a): The temperature-dependence of the pitch length of the HB-CLC; (b): the temperature-dependence of the transmission spectra of cell G; (c): the images of cell G obtained under a polarizer at 165, 175 and 190 °C, respectively.

adjusted within a certain temperature range. This represented a different approach from that of our earlier study,[32,33] in which the prepared Ch-LC film can reflect both right- and left-circularly polarized light (R-CPL and L-CPL) within the same reflection band in a wide temperature range. We believe this is due to the permanent solid helical structure of the polymer network and the nearly temperature-independent pitch length of the bulk LCs with R1011 as chiral dopant in the study. Fig. 6(a) shows the temperature-dependence of the pitch length of the HB-CLC, which is determined by a Cano-rings method taken by a polarized optical microscope (POM, Olympus BX51) at different temperature. It is obvious that the pitch length of the HB-CLC decreases with an increase of the temperature, which indicates that the helical twisting power of the HB-CLC increases with the temperature increasing. Herein, cell G with a double-handed circularly polarized light reflection band was obtained by using the above technique process. In this process, the polymer template with a right-handed helix was originated from mixture 7 and the above HB-CLC with a left-handed helix was infiltrated into the polymer template at a certain temperature. Fig. 6(b) shows the temperature-dependence of the transmission spectra of cell G, it can be seen that the reflection intensity of cell G approaches 100% at 165 °C (curve 1), which means that both red-colored L-CPL and R-CPL have been reflected within the same reflection band by cell G at 165 °C. When the temperature was raised to 175 °C, we were able to observe two reflection bands in the spectra of cell G (curve 2) owing to the difference in the pitch lengths of the polymer template and the bulk LC. This reveals that the single-layer Ch-LC film can reflect green-colored L-CPL and red-colored R-CPL (curve 2) due to the decrease of the pitch length of the HB-CLC with increasing temperature. Furthermore, as the temperature reaches 190 °C, we can also observe two reflection bands in the spectra of the cell G (curve 3), and blue-colored L-CPL and red-colored R-CPL were reflected by cell G. Correspondingly, the images of cell G obtained at these three temperatures under a polarizer were shown in Fig. 6(c). The polarizer used here has two windows for both left-handed polarized light and right-handed polarized light, which is prepared by stacking multi-layer PVA films on TAC substrate, and followed by dyeing and uniaxial tension process. It can be seen that cell G exhibits different reflection modes within the left-handed polarizer and right-handed polarizer windows at 165, 175 and 190 °C, which is in accordance with the above spectrum characteristics. What's more, the above process is a reversible process with an increase and decrease of temperature. Additionally, the transition temperature can be lowered significantly when the above HB-CLCs are used as chiral dopant to add nematic LCs, which means that temperature-tunable PBGs of Ch-LCs with a double-handed circularly polarized light reflection band can be achieved at a lower temperature.

Lasing of a single-layer Ch-LC film with a double-handed circularly polarized light reflection band

It is well known that Ch-LCs are particularly interesting one-dimensional PC materials because of their spontaneous self-assembly into periodic structures, and the PBG can be tuned over a broad range of frequencies, this makes them promising photonic devices, making use of lasing at the PBG edge, and examples have been reported for low-molecular-weight Ch-LCs and for polymeric Ch-LCs.[6–12] In the study reported here, we constructed a single-layer Ch-LC film doped with a lasing dye, which is comprised of the permanent solid helical structure of the polymer template and the bulk liquid crystals (LCs) with the opposite helicity sense, therefore resulting in modulation of reflectance over 50% and lasing emission efficiently. Herein, cell H with a double-handed circularly polarized light reflection band was achieved by using the polymer template with a left-handed helix originated from mixture 1 and mixture 8 with a right-handed helix, in which the composition of mixture 8 was 83.5/16.0/0.5 in wt % (SLC-1717/R811/laser dye, LD) and the chemical structure of the LD is shown in Fig. 1. As shown in Fig. 7(a), the reflection intensity of cell H obtained from the polymer template approaches about 100% (curve 1). That is, both R-CPL and L-CPL had been reflected within the same reflection band by cell H. In the usual single-Ch-LC laser system, it is difficult to obtain relatively high gain with low threshold for lasing, because the PBG is defined only for the circularly polarized light of one handedness. This means that nearly half of the emission from the excited molecules cannot be incorporated in the lasing action.[34] However, in our study, the high reflectance results from Ch-LC film which reflects both R-CPL and L-CPL

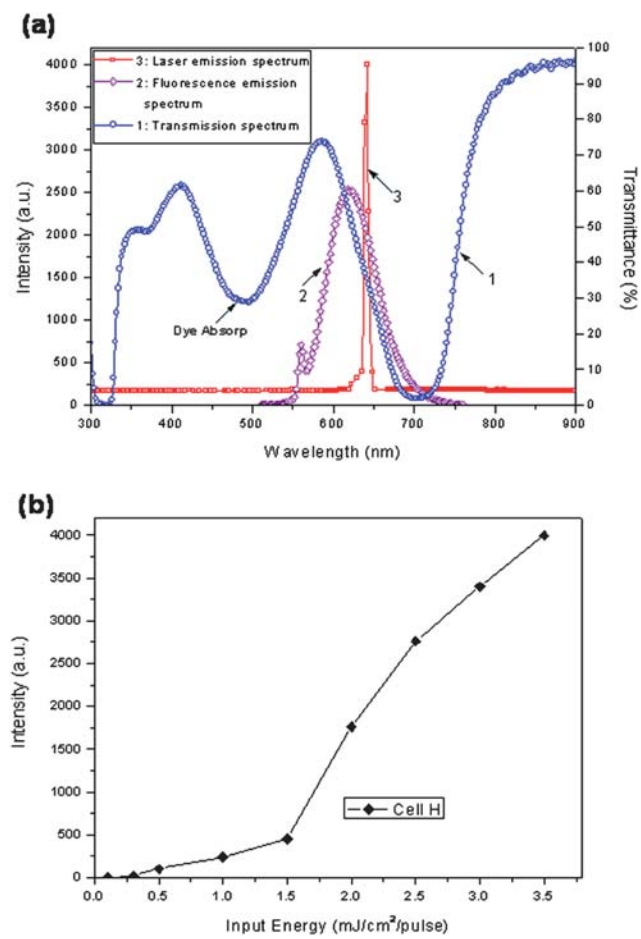

Fig. 7 (a): Transmission spectrum (1), fluorescence emission spectrum (2) and laser emission spectrum (3) by excitation with a second harmonic light of Nd-YAG laser of cell H; (b): the dependence of the emission intensity on the excitation light intensity.

simultaneously within the same reflection band, which indicates that circularly polarized light of both types of handedness can feel the photonic band. Herein, as shown in Fig. 7(a), the Ch-LC film obtained here provides a highly efficient lasing condition in comparison with conventional dye-doped Ch-LCs mirrorless laser assemblies,[6–12] although only one sharp lasing mode is observed at the high-energy PBG edge (curve 3). This is because the reflectance spectrum of the PBG (curve 1) is deviated from the fluorescent emission band (curve 2). On this condition, since the high-energy band edge coincides with the emission peak wavelength, lasing due to the photonic edge occurs at the high-energy band edge. If the low-energy band edge coincided with the emission peak wavelength, lasing at the low-energy band edge would be expected. Fig. 7(b) shows the dependence of the emission intensity on the excitation light intensity. When the light intensity exceeded the pumping energy of 1.5 mJ/pulse, the emission intensity was significantly enhanced. This indicates that the pumping threshold energy for lasing is approximately 1.5 mJ/pulse.

Patterning of a single-layer Ch-LC film with a double-handed circularly polarized light reflection band

The polymer template technique in our study offered a novel opportunity to form a pattern in a single-layer Ch-LC film. To achieve this purpose, mixtures 1 and 9, as shown in Table 1, were used to fabricate the cell with a double-handed circularly polarized light reflection band, in which the helical structure of mixture 1 was left-handed, as mentioned above, and the helical structure of mixture 9 was right-handed. Fig. 8(a) shows the transmission spectra of a cell with mixture 1 before and after UV irradiation, a cell with mixture 9, and cell K. For cell K, two reflection bands in the spectra of the cell (curve 4) were observed, which reflects red-colored L-CPL and green-colored R-CPL, respectively. In order to obtain patterned optical layers, the cells with mixture 1 were irradiated by UV light under masks as shown in the insets of Fig. 8(b) and 8(c). Following that, these two polymer templates obtained by the above process were infiltrated with mixture 9. The images of the cells under a polarizer are shown in Fig. 8(b) and 8(c), it can be seen that both the cells exhibit distinct boundaries and high image contrast. That is, the pattern can only be observed by the left-handed polarizer window; while the whole background can be seen by the right-handed polarizer window. The result could be used in many potential applications including security technologies making use of patterned, monolithic optical layers for both counterfeit deterrence in banknotes and secure documents, and for authentication processes of brand and product protection.

Conclusions

In summary, a novel single-layer Ch-LC film with multi-pitched photonic structure as new PBG materials were fabricated by utilizing a polymer template and various types of Ch-LCs, in which the polymer template was originated from the polymer network of the initial polymer-stabilized cholesteric liquid crystals. The result demonstrated that the helix-structure memory effect by the presence of the polymer template plays a key role in

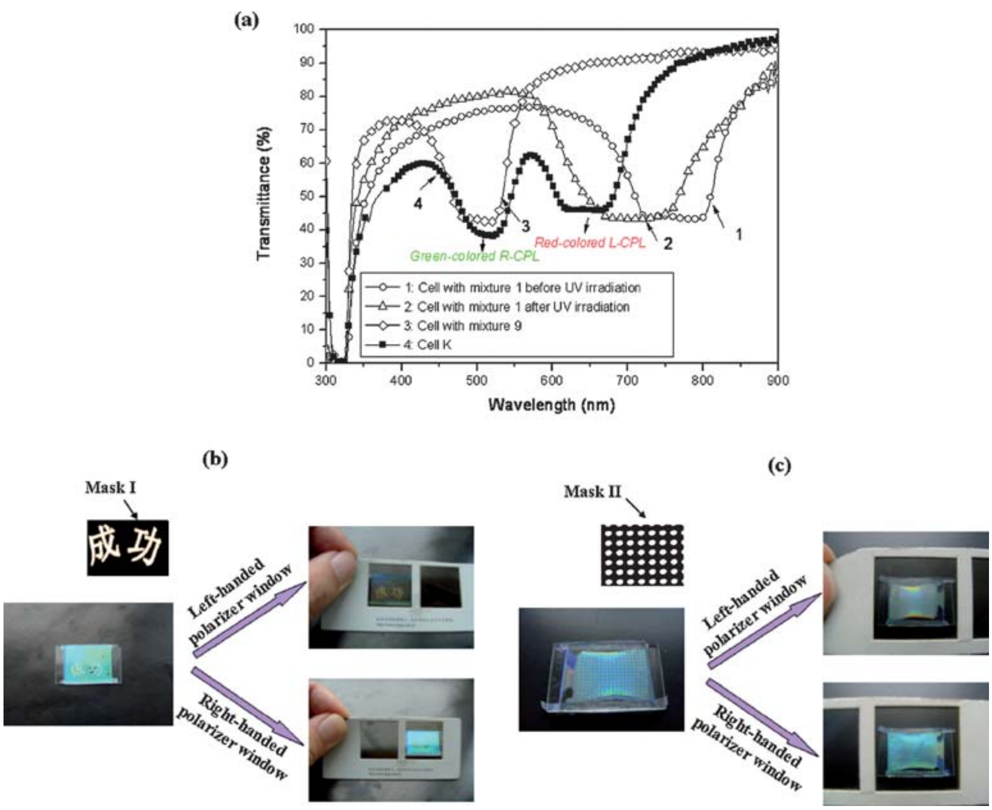

Fig. 8 (a): The transmission spectra of a cell with mixture 1 before and after UV irradiation, a cell with mixture 9, and cell K; (b): the images of the cell obtained by mask I under a polarizer; (c): the images of the cell obtained by mask II under a polarizer.

the development of these novel Ch-LC materials. Simultaneous red, green and blue reflection (multiple PBGs) and wide-band reflection in a single-layer Ch-LC film, temperature-tunable Ch-LC films with a double-handed circularly polarized light reflection band, the lasing emission of the single-layer Ch-LC film, and the patterning of the single-layer Ch-LC film with a double-handed circularly polarized light reflection band can be achieved. The novel Ch-LC composites can be considered as promising materials for photonic and optical applications.

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References

1 E. Yablonovitch, *Phys. Rev. Lett.*, 1987, **58**, 2059.
2 J. D. Joannopoulos, P. R. Villeneuve and S. Fan, *Nature*, 1997, **386**, 143.
3 J. D. Joannopoulos, R. D. Meade and J. N. Winn, *Photonic Crystals*, Princeton University Press, Princeton, NJ, 1995.
4 K. Sakoda, *Optical properties of Photonic Crystals*, Springer-Verlag, Berlin, 2001.
5 A. Stein, F. Li and N. R. Denny, *Chem. Mater.*, 2008, **20**, 649.
6 Y. Katsumi, O. Yutaka, F. Akihiko and O. Masanori, *Japanese Journal of Applied Physics*, 2007, **47**, 5655.
7 S. M. Morris, P. J. W. Hands, S. Findeisen-Tandel, R. H. Cole, T. D. Wilkinson and H. J. Coles, *Opt. Express*, 2008, **16**, 18827.
8 M. Ozaki, Y. Matsuhisa, H. Yoshida, R. Ozaki and A. Fujii, *Phys. Status Solidi A*, 2007, **204**, 3777.
9 T. Manabe, K. Sonoyama, Y. Takanishi, K. Ishikawa and H. Takezoe, *J. Mater. Chem.*, 2008, **18**, 3040.
10 W. Cao, A. Muñoz, P. Palffy-Muhoray and B. Taheri, *Nat. Mater.*, 2002, **1**, 111.
11 M. F. Moreira, I. C. S. Carvalho, W. Cao, C. Bailey, B. Taheri and P. Palffy-Muhoray, *Appl. Phys. Lett.*, 2004, **85**, 2691.
12 H. Finkelmann, S. T. Kim, A. Munoz, P. Palffy-Muhoray and B. Taheri, *Adv. Mater.*, 2001, **13**, 1069.
13 S. S. Choi, S. M. Morris, W. T. S. Huck and H. J. Coles, *Adv. Mater.*, 2009, **21**, 3915.
14 G. Chilaya, in Chirality in Liquid Crystals, *Springer Series Partially Ordered Systems*, ed. H. Kitzerow and C. Bahr, Springer, New York, 2001.
15 P. G. de Gennes and J. Prost, *The Physics of Liquid Crystals*, Clarendon Press, Oxford, 1993.
16 S. Y. Lu and L. C. Chien, *Appl. Phys. Lett.*, 2007, **91**, 131119.
17 A. Bobrovsky and V. Shibaev, *J. Mater. Chem.*, 2009, **19**, 366.
18 R. A. M. Hikmet and R. Polesso, *Adv. Mater.*, 2002, **14**, 502.
19 M. Brehmer, J. Lub and P. V. D. Witte, *Adv. Mater.*, 1998, **10**, 1438.
20 W. Hu, H. Cao, L. Song, H. Y. Zhao, S. J. Li, Z. Yang and H. Yang, *J. Phys. Chem. B*, 2009, **113**, 13882.
21 W. Hu, H. Y. Zhao, L. Song, Z. Yang, H. Cao, Z. H. Cheng, Q. Liu and H. Yang, *Adv. Mater.*, 2010, **22**, 468.
22 D. J. Broer, J. Lub and G. N. Mol, *Nature*, 1995, **378**, 467.
23 D. J. Broer and G. N. Mol, *Adv. Mater.*, 1999, **11**, 573.
24 Z. Y. Bian, K. X. Li, W. Huang, H. Cao, H. Q. Zhang and H. Yang, *Appl. Phys. Lett.*, 2007, **91**, 201908.
25 R. A. M. Hikmet and H. Kemperman, *Nature*, 1998, **392**, 476.
26 B. Fan, S. Vartak, J. N. Eakin and S. M. Faris, *Appl. Phys. Lett.*, 2008, **92**, 061101.
27 H. Yang, K. Mishima, K. Matsuyama, K. I. Hayashi, H. Kikuchi and T. Kajiyama, *Appl. Phys. Lett.*, 2003, **82**, 2407.
28 D. J. Dyer, U. P. Schröder, K. P. Chan and R. J. Twieg, *Chem. Mater.*, 1997, **9**, 1665.
29 M. Mitov, E. Nouvet and N. Dessaud, *Eur. Phys. J. E*, 2004, **15**, 413.
30 S. Relaix, C. Bourgerette and M. Mitov, *Appl. Phys. Lett.*, 2006, **89**, 251907.
31 M. Mitov and N. Dessaud, *Nat. Mater.*, 2006, **5**, 361.
32 J. B. Guo, H. Cao, J. Wei, D. W. Zhang, F. Liu, G. H. Pan, D. Y. Zhao, W. L. He and H. Yang, *Appl. Phys. Lett.*, 2008, **93**, 201901.
33 J. B. Guo, H. Yang, R. Li, N. Ji, X. M. Dong, H. Wu and J. Wei, *J. Phys. Chem. C*, 2009, **113**, 16538.
34 M. H. Song, B. Park, K.-C. Shin, T. Ohta, Y. Tsunoda, H. Hoshi, Y. Takanishi, K. Ishikawa, J. Watanabe, S. Nishimura, T. Toyooka, Z. Zhu, T. M. Swager and H. Takezoe, *Adv. Mater.*, 2004, **16**, 779.
35 J. Hwang, M. H. Song, B. Park, S. Nishimura, T. Toyooka, J. W. Wu, Y. Takanishi, K. Ishikawa and H. Takezoe, *Nat. Mater.*, 2005, **4**, 383.
36 N. Y. Ha, Y. Ohtsuka, S. M. Jeong, S. Nishimura, G. Suzaki, Y. Takanishi, K. Ishikawa and H. Takezoe, *Nat. Mater.*, 2008, **7**, 43.
37 N. Y. Ha, Y. Ohtsuka, S. M. Jeong, S. Nishimura, G. Suzaki, Y. Takanishi, K. Ishikawa and H. Takezoe, *Adv. Mater.*, 2008, **20**, 2503.
38 I. Dierking, *Adv. Mater.*, 2000, **12**, 167.
39 I. Dierking, L. Kosbar, A. Afzali-Ardakani, A. C. Lowe and G. A. Held, *J. Appl. Phys.*, 1997, **81**, 3007.
40 D. J. Broer, J. G. Boven and N. Mol, *Makromol. Chem.*, 1989, **190**, 2255.
41 J. B. Guo, J. Sun, K. X. Li, H. Cao and H. Yang, *Liq. Cryst.*, 2008, **35**, 87.
42 W. L. He, T. Liu, Z. Yang, D. Y. Zhao, W. Huang, H. Cao, G. J. Wang and H. Yang, *Chin. Chem. Lett.*, 2009, **20**, 1303.
43 D. K. Yang, L. C. Chien and J. W. Doane, *Appl. Phys. Lett.*, 1992, **60**, 3102.
44 R. Q. Ma and D. K. Yang, *Phys. Rev. E: Stat. Phys., Plasmas, Fluids, Relat. Interdiscip. Top.*, 2000, **61**, 1567.
45 D. J. Broer and I. Heynderickx, *Macromolecules*, 1990, **23**, 2474.
46 T. Kato, N. Mizoshita and K. Kishimoto, *Angew. Chem., Int. Ed.*, 2006, **45**, 38.

