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Invited Review

Towards nanoscale molecular switch-based liquid crystal displays

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ABSTRACT

Cholesteric liquid crystals (Ch-LCs) have been extensively studied due to their unique self-organized helical molecular structures and selective Bragg reflection properties, which exhibit great potentials for color displays and other practical applications. When functional, nanoscale molecular switches are doped in liquid crystals (LCs), the phases of the LCs or the molecular structures of the Ch-LCs can be changed upon the influence of external stimuli, such as light and temperature. In this paper, the photoresponsive molecular switch-based LCs for display applications are reviewed. The progress and effort in developing molecular switches, the principles of light-tuning, photo-addressed color displays, information processing, bistable displays and flexible displays are presented.

1. Introduction

Liquid crystals (LCs) are fascinating soft materials which exhibit liquid-like fluidity and crystal-like molecular order [1,2]. LCs are optically and dielectrically anisotropic; that is, the physical properties along the directions parallel and perpendicular to the LC molecule are different [3–6]. Due to LCs' anisotropic properties, LC molecules can respond easily to various external stimuli such as electric field, magnetic field, optical field, mechanical force, chemistry, and temperature. Liquid crystal display (LCD) is one of the most important practical applications of LCs, which is the most popular flat panel display (FPD) technology nowadays [7–10]. Transmissive LCDs operated in twisted nematic (TN) [11,12], multi-domain vertical alignment (MVA) [13–19] and in-plane switching (IPS) modes [20] are widely used in LCD industry. For reflective LCDs, the direct-view and projection displays are well developed by exploiting the mechanisms of phase retardation, polarization rotation, Bragg reflection, light absorption and light scattering [21,22]. Cholesteric liquid crystals (Ch-LCs) are extensively studied for reflective displays because of their Bragg reflection properties. In this review, the developments of nanoscale molecular switch-based Ch-LC displays, including light-driven, photo-addressed displays, information processing, bistable and flexible displays are discussed.

2. Molecular switches in LCs

For Ch-LCs, the reflection light wavelength λ from the helical structure is described by Bragg's law: $\lambda = np$ where n is the average index and p is the pitch length of the Ch-LC (the distance of the helical structure undergoing a 360° twist, see Fig. 1). To obtain Ch-LCs, one method is using LCs exhibiting cholesteric phase in the bulk such as cholesteric derivatives [23,24], while the other method is doping chiral dopants into a nematic LC hosts to form helical structures in the mixed systems. The chirality of dopants will transfer into the hosts to form the chiral nematic phase, i.e. Ch-LC phase. The ability of a chiral dopant to twist the nematic host is referred to as helical twisting power (*HTP*), which is defined by $HTP = 1/pc$, where c is the concentration of the chiral dopant [25–27].

If the HTP or p of the Ch-LC can be dynamically controlled, the selective reflection light from the Ch-LC can be tuned according to Bragg equation. The change in *HTP* can be achieved through applying external stimuli such as electric field, thermal energy and chemical reaction to extend, compress, unwind or tilt the helical structure. However, the reflection peaks are usually broadened or decreased, and sometimes scattering light is induced upon the application of external fields [28–30]. An increasingly interesting approach to tune the reflection is by light, which overcomes some disadvantages induced by electric fields, and exploits more opto-optical and electro-optical applications [31–46]. The benefits of photo-driving include easy addressability, fast response and homogeneous reflection peak change, which are desirable features for many practical applications. The dopants consist of chiral agents and photoresponsive moieties, such as spiropyrans, diarylethenes, fulgides and overcrowded alkenes [47–60].

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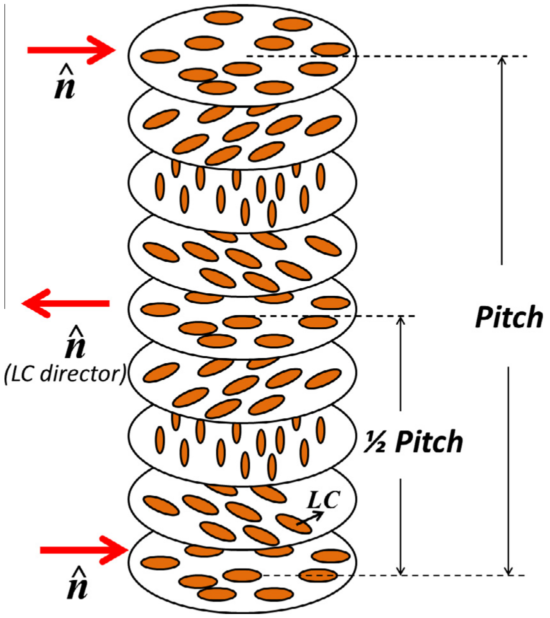

Fig. 1. LC director configuration of Ch-LC.

Azobenzene compounds can be used as photoresponsive materials or chiral dopants [61–75]. Because of the *trans–cis* isomerization of the chiral materials, the molecular configurations of the chiral dopants can be changed by light. The capabilities of the chiral dopants to twist the nematic hosts (i.e., the *HTP* values) are dynamically changed, and then, according to the Bragg condition, the reflection light from the Ch-LC is photo-tuned accordingly [76–83]. The dramatic changes of the HTPs by light can realize more novel color reflective displays.

2.1. Photo-switchable LC phases

When the molecular switches are used as chiral dopants in nematic LCs, they can bring photo-switchable properties and lead to significant changes in the molecular structures or LC phases [84–87]. For instance, it has been shown that photoracemic binaphthanyl derivative could undergo light-initiated conversion from NLCs to Ch-LCs, but not the reverse transition from Ch-LCs to NLCs [88]. Later, a photo-responsive thioxanthene-based guest molecule containing donor and acceptor substituents was used to mix with NLCs [52]. In the *trans* isomer of this guest, a dimethylamono-arene donor moiety is close to the naphthalene chromophore, while in the *cis* isomer, the naphthalene chromophore is facing a nitro-arene acceptor moiety. It was found that conversion of the Ch-LC state I with this guest to the NLC through 313 nm light and another conversion transition from the NLC to the Ch-LC state II through 435 nm light could be achieved, as indicated in Fig. 2. The tuning of the pitch change of the Ch-LC was also obtained reversibly with 365 nm and 435 nm light, and such modulation was stable during 8 switching cycles [52]. An achiral indole fulgide and a chiral binaphthyl ether doped in the NLC also achieved reversible photo-induced change in the Ch-LC due to ring opening and ring closing of the fulgide [89], and bisnaphthol-based chiral fulgide derivatives showed the pitch change of the Ch-LC by photo-chemically switching the fulgide too [90]. However, the fatigue resistance was not high enough so that such change would degrade with switching cycles. From these examples, we can see that the switching between Ch-LC and NLC was dominant in the early stage of the molecular switches used in LCs. The switches between different LC phases can be used for light shutters, optical sensors or displays, etc. [91].

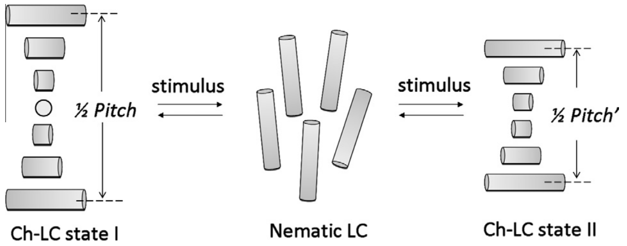

Fig. 2. Switching between the Ch-LC phase and NLC phase by an external stimulus.

2.2. Color change in Ch-LCs

The color change in Ch-LCs has been observed through the change in temperature or the presence of trace amounts of dissolved gases long time ago [92]. Pitch change was then reported by photo-chemical decomposition of cholesterol derivatives [93]. On the one hand, the chiral dopant induces helical structures in the nematic LCs. On the other hand, the photo-isomerization induces reflection color change in the visible range with UV light. The *trans* form can be converted to the *cis* form by using an appropriate UV light wavelength (200–400 nm). The molecule can be converted back to the stable *trans* form with a different wavelength (visible light > 400 nm) or the molecule will thermally relax to the *trans* form. One of such examples is that the chiral molecular switch with the photo-isomerizable double-bond structure was mixed as a dopant with the nematic LC, which brings the opportunity for color display applications [94]. Because the typical length of the molecular switch is 10–80 nm, the molecular switches fall in the nanoscale range. When the UV irradiation was stopped, a reversed thermal helix inversion of the molecular switch occurred after 45 min. The color change with time of the LC E7 with 6.8 wt% molecular switch upon UV irradiation and thermal relaxation are shown in Fig. 3. The process can be repeated over 20 times. With this concept, the studies of molecular switch-based Ch-LC crystal color displays are becoming more and more.

It is highly desirable to dynamically phototune the reflection color over the entire visible spectrum or the primary red, green and blue colors from Ch-LCs with a small dosage of the light-driven chiral molecular switch, since a high concentration of the chiral dopant can often lead to phase separation, coloration, or alter the desired physical properties of LC hosts. This requires the dopants to have high HTP as well as a significant difference of HTP among the various states of the switch. To date, reports on full range color control in induced Ch-LCs usually include helical chiral overcrowded alkenes [95,96], planar chiral azobenzenophanes [53], axially chiral binaphthyl azobenzenes [82,97] and binaphthyl azobenzenes with axial and tetrahedral chirality. However, the color change is either irreversible, or requires a relatively long thermal relaxation. Some systems need a high concentration of planar chiral azobenzenophanes (12 wt%) or binaphthyl azobenzenes with axial and tetrahedral chirality (15 wt%).

Fig. 4 shows high performance photo-reversible full visible light range tuning using an azo-type molecular switch in a small dosage [76]. In the above example, the chiral molecular switch by 6.5 wt% was mixed into NLC without any phase separation exhibited uniform reflection color (Fig. 4). The red-shift by UV light and the reversible color change from red to blue have been achieved by light. The all-optical control overcomes the slow response of thermal relaxation of the molecule itself.

2.3. Critical parameters of molecular switches in LCs

As mentioned above, several critical parameters have to be considered when designing molecular switches in LCs. For the

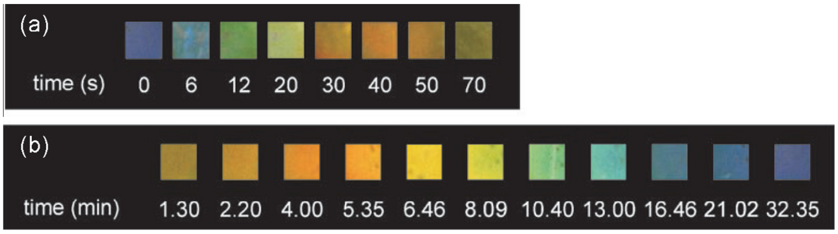

Fig. 3. Color change with time of a LC with a photo-responsive molecular switch: (a) upon UV irradiation (365 nm); and (b) by thermal helix inversion. Reproduced with permission from Ref. [94]. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

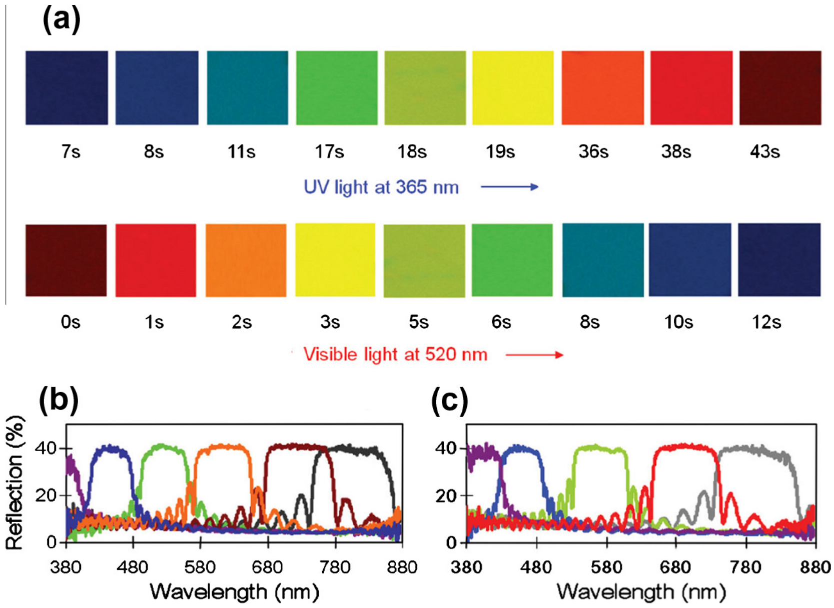

Fig. 4. Full range color phototuning of 6.5 wt% photo-driven molecular switch (azo-type) in LC in a planar cell: (a) reflective polarized optical microscopic colors under UV light (365 nm) and visible light (520 nm) with different time; (b) reflection spectra with UV light (365 nm) irradiation time 3, 8, 16, 25, 40, 47 s (from left to right); and (c) reflection spectra with visible light (520 nm) irradiation time 2, 5, 9, 12, 20 s (from right to left). Reproduced with permission from Ref. [76]. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

phototunable molecular switches, the materials need to be chemically and thermally stable when photo-isomerizable groups are introduced. The chirality of the molecular switches is required to transfer into the nematic LC hosts efficiently, and the dosage should be small to avoid phase separation and coloration. The switching time in the desired region should be in a reasonable range. The transition of the molecular switches in LCs should be reversible. The fatigue resistance needs to be high from the viewpoint of practical applications. These should be considered when designing the molecular switches (in LCs). In the following section, we mainly focus on display applications.

3. Display applications

3.1. Photo-addressed Ch-LC displays

Since the color change can be achieved (some examples are shown in Section 2), the molecular switch-based Ch-LCs show the potential for display applications. Full-color images on solid films of dicholesteryl esters were demonstrated first in thermal mode, and later azobenzene derivative was mixed with dicholesteryl to realize rewritable full-color in the photo mode [61,62,98,99]. Due to the Ch-LC phase in the temperature range of 87–115 °C, the image formed through a photomask must be performed at a high temperature to achieve the Ch-LC phase, then followed by rapid cooling to retain the images in the solid film. The rapid cooling process can prevent crystallization and preserve the Ch-LC structure of the molecules to maintain the Bragg reflection colors. Fig. 5 shows the images on such super-cooled solid film from the Ch-LC phase at a certain temperature range [61].

It is highly desirable more if the Ch-LCs can avoid the heating-up or cooling-down process. Therefore, Ch-LC with photo-responsive dopants at room temperature can make them practical in displays. Efforts are being made for this purpose. To form more promising dopants, two chiral diols, isosorbide and isomannide or similar ideas were chosen to be used in LCs [81]. With different designs, the HTP might be decreased or increased by the UV light. The example of displays at room temperature is shown in Fig. 6, where the pixel-patterned red, green and blue colors can be formed in one cell through the gray photomask [81]. The reversed HTP change can occur in the dark after 48 h due to thermal relaxation of the molecular switch or by the visible light irradiation, which indicates the opportunity of color shift in the whole visible region.

Another example of Ch-LC displays with photo-driven molecular switch at a small dosage which exhibits reversible phototuning across the entire visible region [76] is shown in Fig. 7. Fig. 7a and b shows the primary colors (RGB) formed in the whole cell or single-layer with different time by the visible light irradiation (520 nm), in which the initial cell on the left was driven to near infrared region by UV irradiation at 365 nm. Fig. 7c shows the images with different backgrounds created by photo-masks. Due to the small

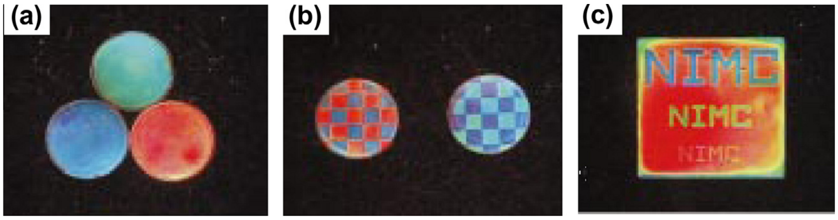

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Fig. 5. Images on a super-cooled solid film from Ch-LC phase with photo-responsive azobenzene derivative through a mask: (a) red, green and blue colors obtained with different UV intensities at 87 °C; (b) images obtained with different temperatures at 246 mJ/cm^2; and (c) images obtained with different optical densities of the photomask at 93 °C and 246 mJ/cm^2. Reproduced with permission from Ref. [61]. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

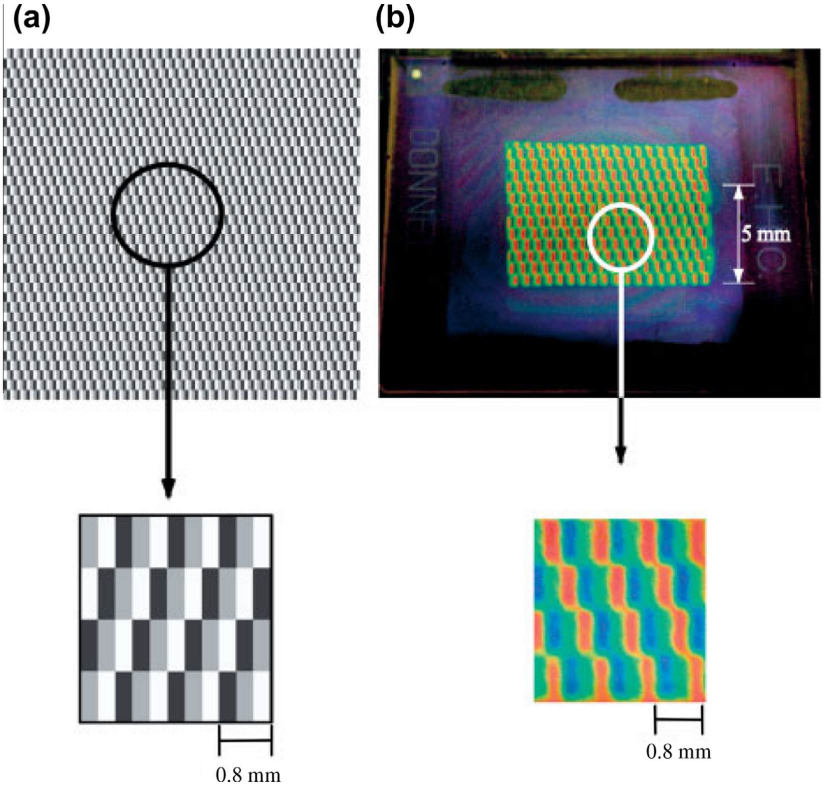

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Fig. 6. Pixel-pattern image achieved by UV light from a Ch-LC with chiral azobenzene dopant: (a) gray mask with different optical densities; and (b) red, green and blue colors obtained by 10 s UV irradiation through the gray mask at 25 °C. Reproduced with permission from Ref. [81]. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

dose doped in LCs, the color and image are more uniform and vivid, and no phase separation in the Ch-LC occurs. The responsive time for the light is fast and the reversible phototuning process can be repeated many times without degradation.

The images on photo-responsive molecular switch-based Ch-LCs can be also written by laser beams [62,98,100–103]. Fig. 8 shows the examples of information recorded by a scanning laser beam in a 2D pattern [100]. The mixture of Ch-LCs with azobenzene nematic LCs demonstrated the capability of phototuning by continuous wave or nanosecond laser beams. In this case, the Ch-LC was driven to photoinduced isotropic state first by UV light and then restored to the Bragg structure by a green laser beam or a CO_2 laser beam. The metastability of the mixture, spatially stable molecular interactions and all-optical applications exhibit the unique application of photo-response materials in LCDs.

3.2. Information processing by using electric effects

There are three textures (states) in Ch-LCs, i.e., planar (P), focal conic (FC) and homeotropic (H) [25]. The textures and the configurations of helical structures are shown in Fig. 9. The planar texture, where the director of the helical axis is perpendicular to the cell surface, can selectively reflect light; that is, its optical state is reflective. If a low electric field or an electric pulse is applied, the focal conic texture is formed. A random distribution of helical axes is the characteristic of the focal conic texture, which scatters the incident light in all directions. If the applied electric field is increased above a threshold value, the focal conic texture is switched into the homeotropic texture, where the helical structure is unwound, and the LC director is aligned in the cell normal direction.

For information processing application, the colored, photo-addressed image driven by light in molecular switch-based Ch-LCs can be hidden by an electric field or electric pulse. Two concepts for hidden information in such Ch-LCs have been used. One way is to apply a high electric field to drive all the LCs to the homeotropic state [104]. Without the helical structure and Bragg reflection, the information is hidden. When the electric field is suddenly removed, the LCs return to the planar state due to the surface boundary condition and the image reappears (Fig. 10a). The other way is

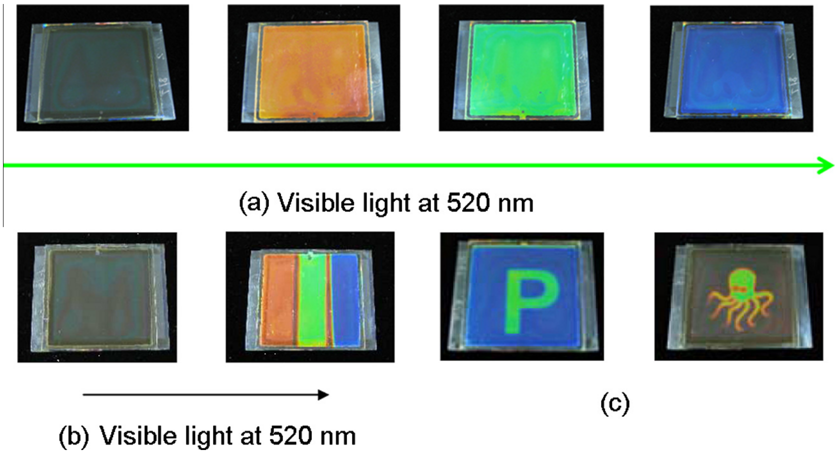

Fig. 7. Colors and images: (a) light-driving of 6.0 wt% photoresponsive dopant in E7 upon visible light (520 nm) with different exposure times; (b) 4.0 wt% dopant in E7 driven by visible light (520 nm) with different exposure times; and (c) photo-addressed displays. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

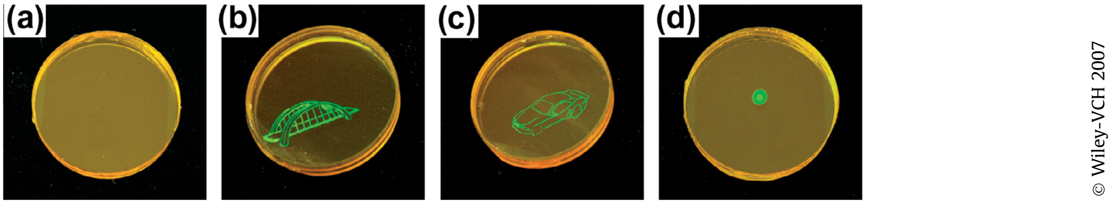

Fig. 8. Images written by laser beams: (a) photoinduced isotropic state of Ch-LC 61 obtained with UV exposure; (b) an image formed by scanning of a focused green laser beam; (c) another image formed by a green laser beam; and (d) a spot formed by a CO_2 laser beam of 10.6 μm wavelength. Reproduced with permission from Ref. [100].

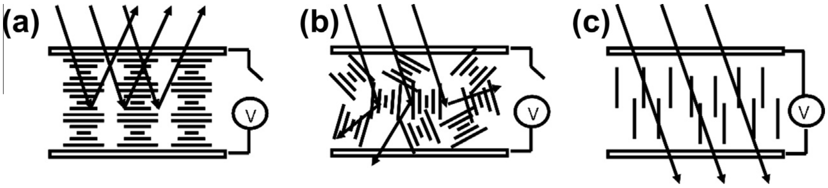

Fig. 9. Textures of Ch-LC: (a) planar; (b) focal conic; and (c) homeotropic texture.

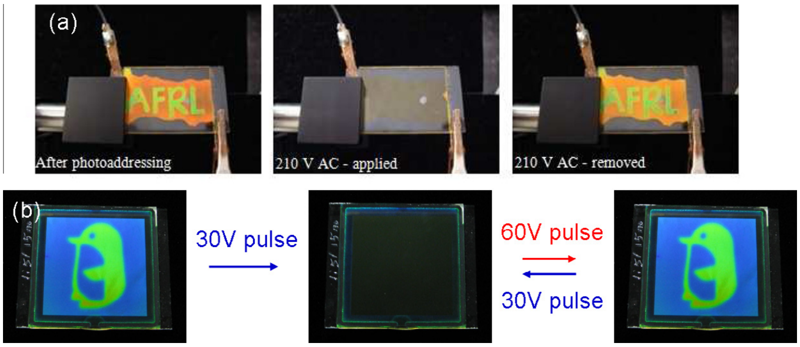

Fig. 10. Molecular switch-based Ch-LCs for information processing: (a) The information recorded in the polymer stabilized Ch-LC (left) can be hidden in the homeotropic state (middle) and reappears by removing the high electric field (right). Reproduced with permission from Ref. [104], Copyright 2010 the Optical Society of America (OSA); and (b) the information in a homeotropic cell (left) was hidden in the focal conic state by a low electric pulse (middle) and reappeared by a high electric pulse (right). Reproduced with permission from Ref. [76]. Copyright, 2010 Royal Society of Chemistry (RSC).

to apply an electric pulse to transform all the LCs into the focal conic state [76]. Due to weak scattering, the image was also hidden. In this case, no electric field was needed to sustain. In order to reproduce the information before the thermal relaxation of the photoresponsive molecular switch, a high electric pulse is used to make the LCs return to the initial planar state (Fig. 10b). Such manipulation for hiding and revealing information processing can be repeated many times until isomerization of molecules via thermal-relaxation occurs. The display of background color and information contents can be controlled by light tuning.

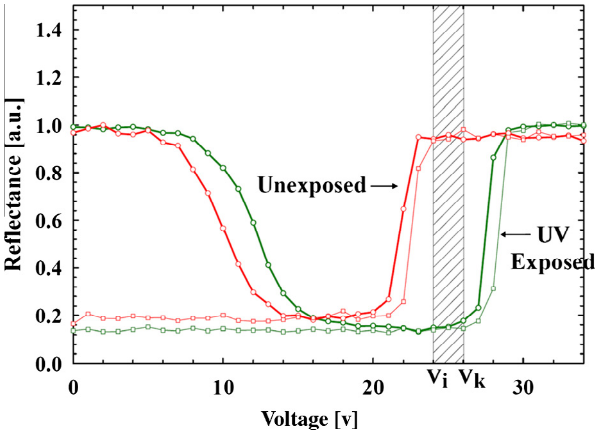

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Fig. 11. Electro-optical curve of a bistable Ch-LC system based the photo-responsive molecular switch concept. Reproduced with permission from Ref. [112].

3.3. Bistable displays

The chiral molecular switch-based Ch-LCs are amenable to the development of electrically-switchable bistable display applications by using polymer stabilized or surface stabilized chiral nematic texture technologies [105–108]. The conventional bistable Ch-LCs have been studied since 1990s. Two kinds of bistable modes, i.e., bistable mode between focal conic state and planar state [108–110] or bistable mode between homeotropic state and planar state [111] have been developed. The bistability based on chiral molecular switches in optically addressed displays can be realized by means of electric driving, whereby the image is retained indefinitely and can be also erased electrically. The image can be thermally stable and be retained indefinitely, through electrically switching either the image or the image background to the focal conic state before it thermally relaxes.

The response of a system of bistable Ch-LC with photo-responsive molecular switch is shown in Fig. 11. In this system, the UV-exposed LC has higher threshold voltage. The image or its background can be electrically selected since the HTP can be changed by the light [112,113]. An advantage of such bistable displays is that a thermally stable image is captured without patterned electrodes or costly electronic drive and control circuitry, and the image retains indefinitely until electrically erased. A phototunable cholesteric layer simply sandwiched between two blank, unpatterned transparent electrodes is enough. For example, optical writing is done in the planar state through a photomask by UV light. The reflective image can be hidden in focal conic texture by applying a low electric pulse (such as 22.5 V) after photo-addressing, as shown in Fig. 11. By applying the electric pulse between V_i and V_k to this image so as to make the UV exposed region in the focal conic texture and the UV unexposed region in the planar texture. Therefore, the optically written image is stored indefinitely since the planar and focal conic textures are both stable (for instance, see Fig. 12a–c). The image and background color in the cell can be adjusted arbitrarily by light.

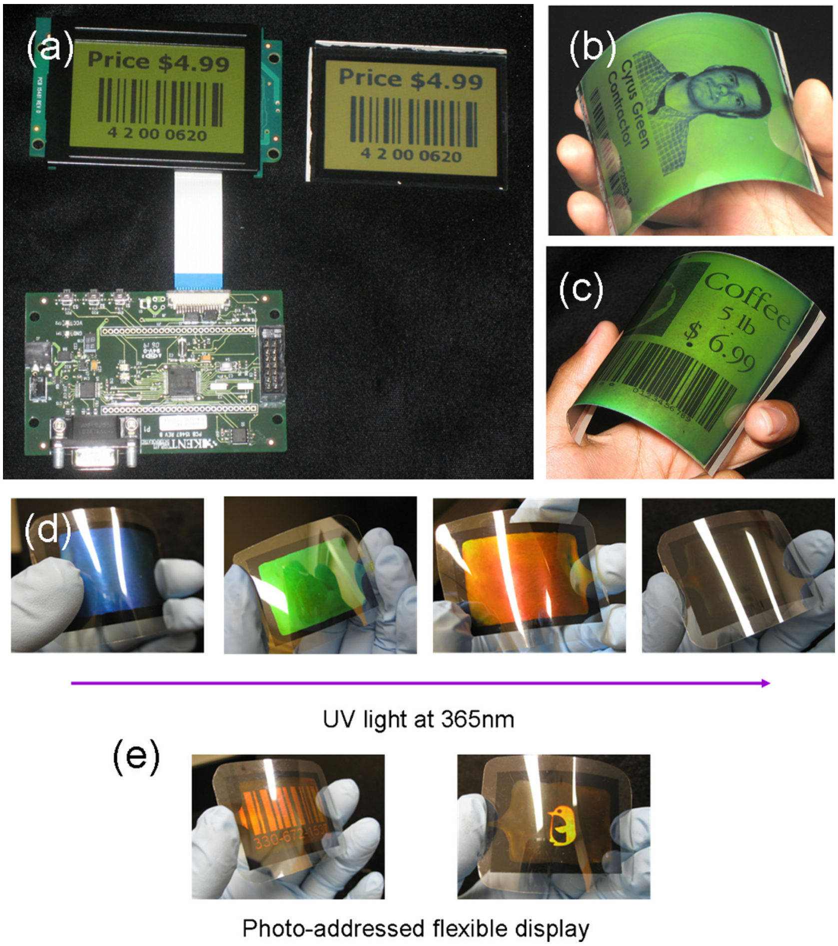

Fig. 12. Flexible displays of molecular switch-based Ch-LCs: (a) no need of complex electric circuitry; (b) and (c) bistable flexible display examples; (a)–(c) Reproduced with permission from Refs. [112] and [113]. Copyright, John Wiley and Sons; (d) colors driven by UV light (365 nm) with different exposure times; and (e) photo-addressed flexible displays.

3.4. Flexible application

Certainly, since LC is a class of "soft" matter, it can be used for flexible displays, which is one of the next-generation display styles [114–117]. Fig. 12 shows examples of applications in flexible displays. Traditionally, to form a sophisticated (bistable) display image, a complex electric circuitry is necessary (Fig. 12a-left) while photo-addressed image can be achieved through light writing methods without any complex electric circuitry (Fig. 12a-right). The flexible cell can be driven by UV light to obtain blue, green, red and transparent (near IR) color (Fig. 12d), and photo-addressed flexible displays can be achieved through photomask (Fig. 12e) or in bistable state (Fig. 12a–c). Again, the advantage of light-driven, photo-addressed molecular switch-based Ch-LC flexible displays does not need complicated driving controls.

4. Conclusions

In conclusions, nanoscale molecular switch-based cholesteric LCD is moving towards practical applications. The photo-responsive dopants bring phototuning ability into the conventional Ch-LC displays. The new materials can be used for light-driven, photo-addressed displays, information processing, bistable and flexible displays. Because of the responsive nature of molecular switches upon the application of external stimuli, the molecular switch-based LCs become a desirable candidate for novel display technologies. The further use of molecular switches in LCs, blue phases, smectic phases or polymers will bring more opportunities for smart materials and device applications.

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