Security devices based on liquid crystals doped with a colour dye

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Liquid crystal properties make them useful for the development of security devices in applications of authentication and detection of fakes. Induced orientation of liquid crystal molecules and birefringence are the two main properties used in security devices.

Employing liquid crystal and dichroic colorants, we have developed devices that show, with the aid of a polarizer, multiple images on each side of the device. Rubbed polyimide is used as alignment layer on each substrate of the LC cell. By rubbing the polyimide in different directions in each substrate it is possible to create any kind of symbols, drawings or motifs with a greyscale; the more complex the created device is, the more difficult is to fake it.

To identify the motifs it is necessary to use polarized light. Depending on whether the polarizer is located in front of the LC cell or behind it, different motifs from one or the other substrate are shown. The effect arises from the dopant colour dye added to the liquid crystal, the induced orientation and the twist structure. In practice, a grazing reflection on a dielectric surface is polarized enough to see the effect. Any LC flat panel display can obviously be used as backlight as well.

Keywords: liquid crystal, security device, dichroic dye, absorption, polarized light, latent image.

1. Introduction

In recent years, liquid crystal displays (LCDs) are used in many kinds of appliances, mobile telephones, notebook PC and TV monitors; they play a major role for the application of flat panel displays. But optical properties of liquid crystals, handled the right way, can also be used to achieve different effects on the light that goes through unplugged passive devices; these effects are easily verifiable with the naked eye or with the help of simple optical devices [1–4].

Taking advantage of LC orientation properties and birefringence, it is possible to fabricate devices with permanent motifs that vary depending on the orientation of the cell or the substrate where the light impinges from. This opens the opportunity of developing a new security and authentication system to be applied in banknotes, checks, document identification, credit cards and other valuable documents in general, based on liquid crystals doped with dichroic dyes.

Liquid crystal twisted nematic (TN) structure is one of the most common in displays. In this work, TN structure with multi-domains is used to create an LC alignment patterning to be applied in optical security devices with no voltage required [5,6]. Applying different alignment conditions on the opposing glass substrates (combined with several photolithographic processes) makes it possible to create different latent images on each side of one single device [7,8]. The alignment patterning is carried out by mechanical rubbing that is used in large and small areas and multi-domain patterning. Varying rubbing directions a pattern of LC alignment in different directions can be created through a relatively simple manufacturing process.

This device allows seeing different motifs with the simple aid of polarized light, generated either from a polarizer, a standard LC display (used as backlight) or even an oblique reflection of a dielectric reflecting surface. By varying the alignment conditions of the glass surfaces that confine the liquid crystal, different easily recognizable drawings can be created, whether saturated monochrome B/W or greyscale [9,10]. The manufacturing system presented in this paper allows us to obtain independent images on both sides of the device. These images are seen in negative when rotating 90 degrees the polarization axis of the light used for verification.

2. Operating principle

The operating principle of the device is based on the ability of a dichroic dye -oriented by the LC- to absorb light polarized parallel to the direction of its long axis [11]. Rubbing treatments promote a homogenous alignment (parallel to the confining plates) of the LC molecules, but with different orientation directions, which identify the areas that will eventually appear clear or dark when polarized light impinges the device.

We have employed a standard manufacturing process commonly used in liquid crystal displays fabrication, where confining glass plates are conditioned to obtain several liquid crystal orientation directions across each surface of one single device.

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Combining orientations on either glass plate, different twist, and parallel alignment regions are defined on the liquid crystal layer doped with dichroic dye. The molecules of the dichroic dye are elongated, allowing the dopant (in small concentrations) to orient parallel to the neighbouring LC orientation. This results in dichroic absorption of the incoming light, being maximum for light polarized parallel to the LC director (hence, parallel to the long axis of the dye), and minimum for the light polarized on any direction lying on the plane perpendicular to the director. Intermediate directions between the long axis and the perpendicular directions give partial absorption that may be used to create a greyscale.

Therefore, every region of the device will absorb the polarized light corresponding to the local alignment that has been induced onto the liquid crystal (and consequently to the colorant) in every specific area. This phenomenon permits the generation of latent images. Different alignment patterns on each surface of one single device allow us to create domains inside the device with twisted nematic structure while other domains adopt parallel structure as shown in Fig. 1.

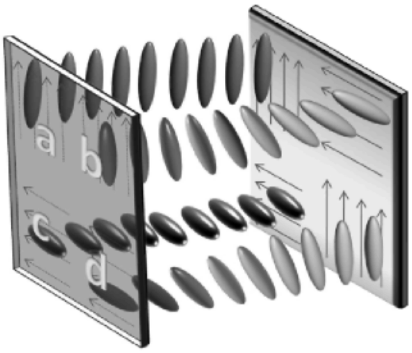

Fig. 1. A sketch of a simple LC security device. Alignment layers have been conditioned in order to induce different orientations on LC molecules generating the desired motifs. Twisted nematic structure allows creating different alignment patterns on each surface of one single device and makes it possible to create multiple latent images in the same device.

The liquid crystal doped with dichroic dye is oriented by the alignment layer on each side of the cell. The molecules rotate (perform a twist) if the alignment direction on either substrates is different in any given area of the cell Figs. 1(b) and (d). In contrast, the molecules do not rotate if the alignment directions are parallel [Figs. 1(a) and (c)]. The orientation of liquid crystal molecules is determined by the selectively rubbed polyimide layer according to the desired motifs.

For a better understanding of the device behaviour we will study eight different situations (in Fig. 2). In the first four cases, light impinging from left is vertically polarized. In Fig. 2(a), vertically polarized light remains vertical and is absorbed by the colorant, obtaining a dark state. In Fig. 2(b), light is rotated by the twist structure following the LC molecules and, in the same way, staying parallel to the dye molecules; light is absorbed by the colorant and a dark state is obtained. In Fig. 2(c), vertically linear polarized light does not rotate and goes out vertically polarized, obtaining a bright state. In Fig. 2(d), light polarization is rotated and travels whole the way perpendicular to the LC and colorant molecules, obtaining a bright state at the exit with horizontally polarized light.

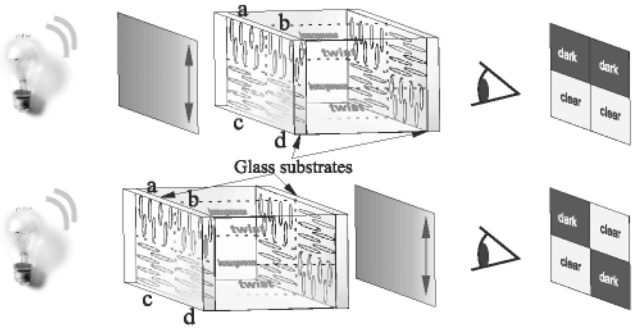

Fig. 2. Schematics of security device operating principle. Dichroic dye absorbs the light depending on orientation of LC molecules and polarization of light. Situation of polarizer determines which of two latent images is visible. Only an image generated on a substrate closest to a polarizer is seen.

If the impinging light were horizontally polarized, the result would be the complementary. So, in the first four cases we can conclude that the output will be bright or dark depending on whether the impinging linear polarizations are respectively perpendicular or parallel to the molecules of the entering surface.

For the other four cases let us consider unpolarized light impinging from left and a vertically linear polarizer placed at the light exit.

In the parallel cases, Figs. 2(a) and 2(c), both components of light will continue in the same direction while in twist cases, Figs. 2(b) and 2(d), both components of light will rotate following the LC molecules. In the four cases, the light component that is parallel to the LC molecules will be absorbed by the colorant so, at the exit of the device (before crossing the linear polarizer) we will have a unique linear polarized light component, horizontal in cases of Figs. 2(a) and 2(d) and vertical in cases of Figs. 2(b) and 2(d). Placing a vertical linear polarizer at the exit, a dark state is obtained for cases of Figs. 2(a) and 2(d) and a bright state for cases of Figs. 2(b) and 2(c). Again, as in the previous four cases, a state shall be clear or a dark depending only on whether the polarizer placed in the corresponding surface is perpendicular or parallel to the LC. This makes it possible to generate different latent images on each face of the device; these images can be seen independently. Figure 2 shows the visual effect that is achieved depending on where the polarizer is placed, either in front of the device or behind it. Entrance surface absorbs light in each case so dark and clear states are generated. Different images are seen in either case.

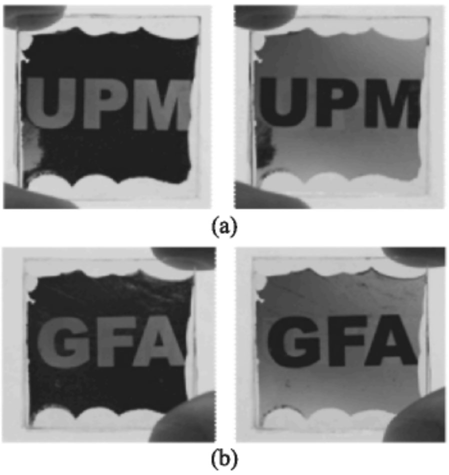

Fig. 3. Pictures of a device when a polarizer is placed in front of the LC cell (a) and when a polarizer is placed behind LC cell (b). Complementary images are obtained when a polarizer is rotated 90°.

Figure 3(a) shows two images visible when a polarizer is placed in front of the LC cell. The second image is shown by rotating the polarizer 90 degrees obtaining the complementary image. If the polarizer is placed behind the LC device, the other set of images corresponding to the other surface will be visible, as shown in Fig. 3(b). If no polarizer is used, no image is visible but a homogeneous colour corresponding to the dichroic dye used. It is not really necessary to use the polarizer to check the latent images; it is enough to watch the device with partially polarized light, for example, from a reflection of the floor or a table with enough grazing angle on a dielectric surface. The decompensation of light components caused by proximity to the Brewster angle allows us to observe the effect. It is also possible to observe the latent images using light outgoing from any liquid crystal display, like those in computers or mobile-phones that could help the massive deployment of these devices as security element in labels or bank notes, for example.

3. Experimental

A commercial dichroic mixture named TedDis 150, based on Merck MLC 6025-000 and doped with small amounts of red, yellow, and blue dyes from Roche has been employed in most of the manufactured devices. The mixture contains a small amount of a chiral dopant to strengthen the twist rotation structure of the liquid crystal, avoiding reverse twist generation.

The alignment layer is polyimide (PIA2000). The deposition of the polyimide alignment layer is made by spin-coating at 2500 rpm for 20 seconds obtaining an average uniform layer thickness of about 200 nm. Polyimide is cured by introducing the samples in a preheated oven at 200°C for 45 minutes before moving on to the stage of rubbing and creating latent images.

Standard rubbing is done using a custom made buffing-machine that performs a rotational-translational motion.

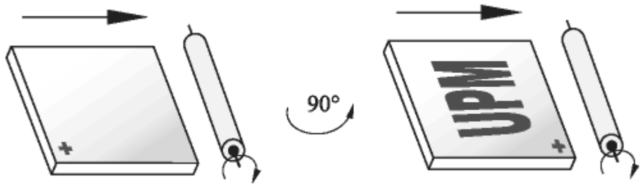

Fig. 4. Schematics of a rubbing procedure that allows "drawing" the latent motifs in a polyimide alignment layer. Firstly, a surface is rubbed in one direction and then, after a photolithographic process, a substrate is rotated 90° to change alignment of uncovered zones by rubbing again while maintaining remaining areas covered by a photoresist.

The rubbing is done in stages, according to the motifs desired to be "drawn" on the alignment surface. Latent images must be aligned independently on each substrate to be seen independently as well. The standard procedure, shown in Fig. 4, requires only two buffing steps. In the first buffing, the substrate as a whole is rubbed in one direction. Then, the sample undergoes a photolithographic process that covers the unwanted surface, and the remaining motifs are rubbed in perpendicular direction.

A standard photolithographic mask is applied to the substrates prior to the above mentioned rubbing as indicated in Fig. 4. A conventional positive photoresist (Rohm & Haas S-1818 G2) is spinned onto the rubbed surface at 4500 rpm obtaining an average thickness of about 1.8 μm. Once cured (90°C, 30 min.), it is irradiated with UV light using masks like those shown in Fig. 5 to create the pattern of the images. After developing the photoresist layer, exposed areas are removed while the rest of them remain covered with photoresist. Covered areas are not affected by the second rubbing process, maintaining the alignment direction induced by the first rubbing (see Fig. 4). Once the rubbing process is complete, the existing photoresist layer is removed with acetone and substrates are ready for assembling. The spatial resolution of the device is determined by the photolithographic processes that precede the rubbing of the substrates.

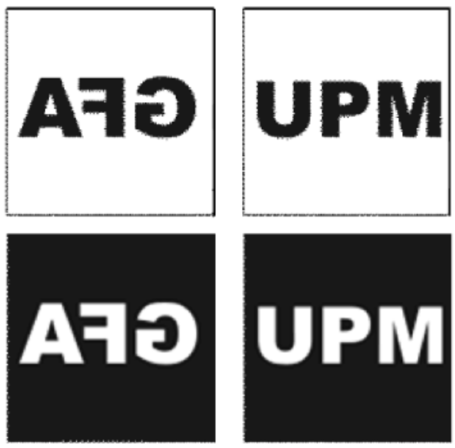

Fig. 5. Pattern design of masks used in a manufacturing process to create desired latent images inducing different alignment directions.

4. Greyscale devices

Using the same principles of bi-directional rubbing, a new rubbing protocol to implement continuous greyscale latent images in LC security devices has been developed. This protocol makes it possible radial and circular (or tangential) rubbing, as shown in Fig. 6, (a1) and (a2), and is based on the design and manufacture of a goniometric gradual-step system and on a circular rotation rubbing equipment. The circular rubbing creates a tangential alignment from the centre. The alignment generated by radial rubbing varies the orientation of the molecules from grade to grade (continuously varying spatial alignment between 0° and 360°) to complete the circle. The alignment direction of LC molecules at any given point is slightly rotated compared to the direction of the molecules at an immediately adjacent point. The result of a circular rubbing is shown in Fig. 6 (b1), it is an easily recognizable greyscale motif displayed in a semicircle. A radial rubbing produces a similar effect that varies continuously through the device but with inverted greyscale since the radial and the circular rubbing are mutually perpendicular at each point [see Fig. 6 (b2)]. At the same time, it is possible to combine the linear and circular-radial alignment techniques discussed above in order to create more sophisticated devices and complex latent images, see Fig. 6, (c1) and (c2).

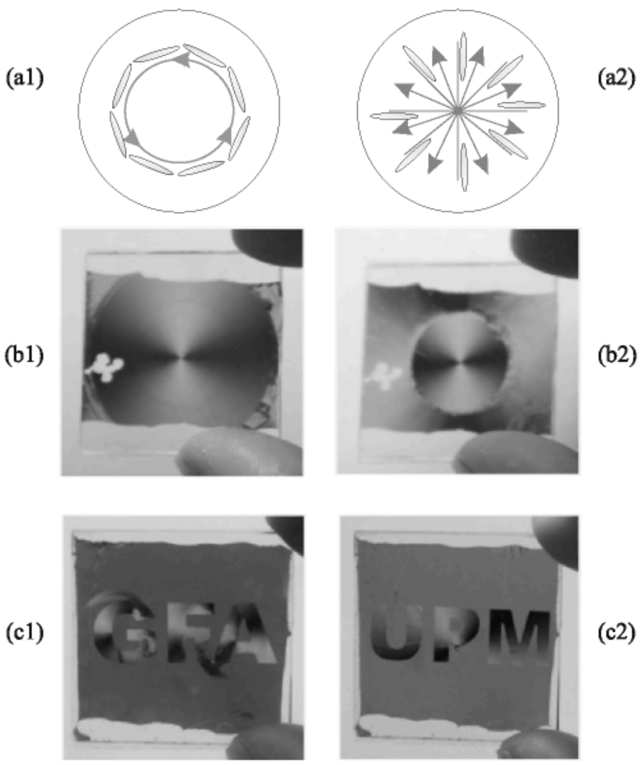

Fig. 6. Schematics of circular (a1) and radial (a2) rubbing direction, LC molecules are oriented according to this alignment. We can achieve continuous greyscales as in picture (b1) which corresponds to circular rubbing, and (b2) which is a combination of radial and circular rubbing. Pictures (c1) and (c2) show a combination of linear standard rubbing and circular rubbing.

The thickness of the fabricated devices (see Fig. 7) is another important issue in manufacturing. The developed devices require employ nematic LC twist configuration. A lower limit in the device thickness is determined by the Mauguin's condition. This condition for the incident light to follow correctly the rotation imposed by the twist of the LC molecules

$$\Delta n \cdot d >> \frac{\Phi \lambda}{\pi}, \tag{1}$$

where d is the thickness of the cell and Φ is the total twist angle. For a standard TN cell, $\Phi = \pi/2$, and the expression reduces to

$$2 \cdot \Delta n \cdot d >> \lambda . \tag{2}$$

The physical meaning of this condition is simple: if there is some difference between the optical paths (nd) of the ordinary and the extraordinary axis of the LC molecules, the linearly polarized light is spread more easily than the elliptical light. Otherwise, the linear polarization is not maintained. For this reason, in this application, "thick" devices are preferred in comparison to those commonly used in LC display manufacturing, where priority is often to reduce the switching time of the liquid crystal by limiting the thickness, employing first or second minimum. The devices shown in this work do not switch, therefore no dynamic limitations exist. We use cylindrical shaped spacers of 14 μm in diameter to ensure sufficient separation between substrates so it works properly.

Manufactured devices show improved contrast ratio and transmission characteristics. Figure 7 shows the transmission for a dark state and a clear state in visible wavelength range. Light absorption when a dark state is obtained (light is polarized in the same direction of LC and dichroic dye long axis) is about 98%.

A large thickness also implies that a lower concentration of colorant can be used, improving the colorant alignment and the behaviour of the device. Large thickness is necessary in these devices, thinner devices would show undefined

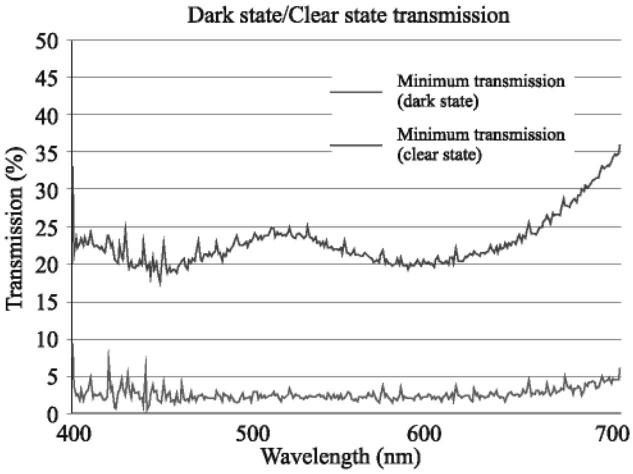

Fig. 7. Transmission obtained for one standard manufactured device in the case of dark state and clear state.

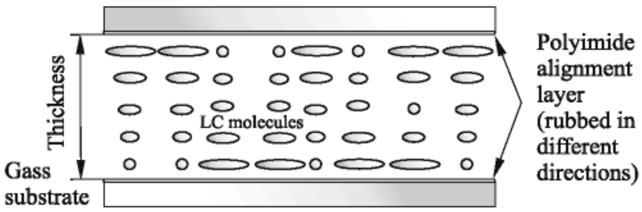

Fig. 8. Simple drawing of a security device LC cell. Glass substrates are coated with the polyimide alignment layer that induces different orientation in both planes to the LC molecules doped with a dichroic dye.

latent images due to the low contrast achieved. Too thinner devices are not really interesting for industrial application. The thicker the device, absorption is higher too and a better contrast ratio is achieved. This allows us to employ liquid crystals with low optical birefringence for the manufacturing.

5. Conclusions

We have developed a device to create latent images for security devices manufacturing. Different dual images can be shown in one single device and they are made visible separately using a linear polarizer. Greyscales have been achieved using new rubbing techniques. We present a simple manufacturing process that allows us to get complex and sophisticated security devices for application in valuable documents, banknotes, credit cards, etc.

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