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Amrita Mukherjee [a], Gyu Hyung Yang [a b], Heon Jeong [a b], Prasenjit Nayek [a], Shin-Woong Kang [a], Seung Hee Lee [a b], Seung Ho Hong [c], Hyuck Jin Lee [c] & Sung-Tae Shin [c]

[a] Department of BIN Fusion Technology, Chonbuk National University, Jeonju, Jeonbuk, Republic of Korea

[b] Department of Polymer Nano-Science and Technology, Chonbuk National University, Jeonju, Jeonbuk, Republic of Korea

[c] LCD Research Center, LCD Division of Samsung Electronics, Giheung, Gyeonggi-do, Republic of Korea

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Emergence of a novel optically isotropic transient state with low frequency in a blue phase liquid crystal mixture

Amrita Mukherjee[a], Gyu Hyung Yang[a,b], Heon Jeong[a,b], Prasenjit Nayek[a], Shin-Woong Kang[a]*, Seung Hee Lee[a,b]*, Seung Ho Hong[c], Hyuck Jin Lee[c] and Sung-Tae Shin[c]

[a]*Department of BIN Fusion Technology, Chonbuk National University, Jeonju, Jeonbuk, Republic of Korea;* [b]*Department of Polymer Nano-Science and Technology, Chonbuk National University, Jeonju, Jeonbuk, Republic of Korea;* [c]*LCD Research Center, LCD Division of Samsung Electronics, Giheung, Gyeonggi-do, Republic of Korea*

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The electro-optical hysteresis characteristics in a blue phase liquid crystal system showing multi-domain platelet type body-centred cubic (BP-I) structure at different levels of applied electric field have been investigated. The BP-I state possesses a large degree of hysteresis due to its compact body-centred cubic structure. The hysteresis behaviour is discussed in the context of simulation and theoretical studies reported earlier. When the cell is driven with a low frequency square wave, a novel optically isotropic state with high value of the Kerr constant has been observed. Hysteresis and response time both show relatively lower values in this state with respect to the multi-domain BP-I state.

Keywords: blue phase; chiral nematic liquid crystal; hysteresis

1. Introduction

Blue phase (BP) liquid crystalline (LC) materials are becoming increasingly significant in new generation liquid crystal display (LCD) devices due to their extraordinary optical properties [1–3]. The BP LCD exhibits much wider viewing angle characteristics than those of traditional LCDs, and it can refresh images on the screen more rapidly than earlier LCDs due to its sub-millisecond grey-to-grey response time [4], significantly reducing blur. Moreover, a BP LCD does not require a LC alignment layer such as a polyimide, consequently eliminating the need for mechanical or chemical alignment processes. Accordingly, it can be made using fewer processing steps than current LCDs, and BP displays are therefore potentially less expensive. In addition, the transmittance of BP LCs is insensitive to the cell gap, provided this exceeds 2–3 μm [5]. This cell gap insensitivity is mainly beneficial for large-panel LCDs in terms of manufacturing yield.

On the basis of Landau and defect theories [6], three distinct thermodynamically stable blue phases are known: BP-I (body-centred cubic structure), BP-II (simple cubic), and BP-III (amorphous with a local cubic lattice structure in the director field). The complex structure of the BP LCs in general limits their temperature range to only a few degrees Kelvin below the clearing point of the material. However, following polymer stabilisation the temperature range of BP LCs can be extended over 60°C [1]. LC quaternary mixtures [7] and chiral additive-doped bent-core LCs [8] can also extend BP range. Furthermore, the stabilising effects of nano-particles on cholesteric BP have also been reported recently [9, 10].

However, in practical display applications practical issues such as operating voltage (V_{OP}), hysteresis, residual birefringence and long-term stability remain to be addressed in both pure and stabilised BP systems. Thermal hysteresis in BP LCs over a variety of heating and cooling cycles has been reported previously by Collings and McColl [11]. The large electro-optical Kerr effect in BP LCs involves voltage-dependent hysteresis, which influences the accuracy of the grey scale level and to some extent restricts its application in devices. Recently this essential issue has been addressed by Chen *et al.* in pure polymer-stabilised BP LC systems for both BP-I and BP-II phases [12]. They have shown that the BP-I phase exhibits a slow response time and a considerable degree of hysteresis. After polymer stabilisation, however, its operating voltage is increased by 15%, but its response time is improved drastically and the degree of hysteresis is reduced.

In the present paper we describe the hysteresis effects in a BP LC system in the BP-I phase for different conditions of applied electric field. The observations on detailed electro-optical behaviour are discussed on the basis of the simulation and theoretical

*Corresponding author. Email: swkang@jbnu.ac.kr (SWK), lsh1@chonbuk.ac.kr (SHL)

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research results previously reported. For certain values of applied electric field a novel optically isotropic state has been observed with high Kerr constant and lower hysteresis.

2. Experimental

A nematic liquid crystal mixture consisting of cyanobiphenyl and cyanobiphenyl groupings was employed, with dielectric anisotropy ($\Delta\varepsilon$) $\sim$ 14 and birefringence (Δn) $\sim$ 0.25, mixed with two chiral dopants, ZLI4572 (8 wt%) and CB15 (22 wt%). The mixture (LC Matter Corporation, USA) showed the following phase sequence: Iso–65°C–BP-I–60°C–N*. The LC mixture was sandwiched between two glass substrates. The upper glass substrate was plain glass, and the lower substrate was coated with patterned indium–tin oxide electrodes with in-plane switching geometry. The cell parameters were as follows: electrode width $\sim$ 4 μm, electrode spacing $\sim$ 4 μm, and cell gap $\sim$ 10 μm. To measure the electro-optical properties of a BP LC sample a white light source was used. The BP LC cells were placed between crossed polarisers and the textures were observed at two different light intensities using an optical polarising microscope (Nikon DXM1200). Before taking a reading the sample was stabilised at BP-I temperature ($\sim$ 62°C). In the IPS cell the electric field-induced birefringence was in the direction lateral to the plane of the substrate. To maximise the transmittance the striped electrode was oriented at 45° to the transmission axis of the polariser. A root-mean-square voltage (V_{rms}) with high (1 kHz) and low (60, 10 Hz) frequency square waves was used to drive the IPS cell. The voltage ramping rate was 50 ms per step ($\sim$ 20 V s^{-1}). The optical transmittance was calculated using an image analyser *i*-solution (*i*M Technology). The transmittance was normalised to that when the BP LC cell was in the isotropic state and the two polarisers were parallel.

3. Results and discussion

Figure 1 shows microscope images of the BP-I state observed under crossed polarisers, with both increasing and decreasing amplitude of applied AC voltage of frequency 1 kHz. The characteristic platelet texture with domains reflecting distinct colours confirmed the BP-I state. The classic shadow-like platelets were observed to have an average domain size of 50 μm.

In Figure 1 it is seen that at about 15 V_{rms} the brightness of the images for ascending (Figure 1(b)) and descending (Figure 1(f)) applied voltages was completely different. From the field-dependent textural observations it is therefore evident that the BP-I phase possessed a significant amount of electro-optical hysteresis. At the highest applied voltage a clear flow effect was observed in the texture. In addition it should be noted that the initial texture of the multi-domain BP-I phase before applying the voltage (Figure 1(a)) was slightly different from the texture after applying one cycle of voltage (Figure 1(g)).

Figure 2 shows the normalised voltage-dependent transmittance (V–T) curves measured in the BP-I phase. Normalisation was achieved by the optical transmittance of the cell in the isotropic phase under parallel polarisers. In the BP-I state, as the voltage increased the transmittance gradually increased, reaching a peak ($\sim$ 70%) at $V_{\mathrm{OP}} = 45$ V_{rms}. As seen in Figure 2, as the voltage is reduced, the transmittance decreases, but in this case taking a different path.

To confirm the hysteresis effect we repeated the voltage sweep over a number of cycles. A shift of hysteresis towards the lower voltage side, as indicated by the arrows, was observed for the next voltage cycles, as shown in Figure 2. The results of hysteresis measurements were consistent with the textural observations during the switching cycles, as demonstrated in Figure 1. The voltage variation at a half-maximum of transmittance between forward and backward directions of applied voltage, are a measure of hysteresis, ΔV is $\sim$ 8 V_{rms} and the ratio $\Delta V/V_{\mathrm{OP}}$ is $\sim$ 17.8% in the sample. For practical display applications this ratio should be negligible, and further research is in hand to reduce it.

Since the BP-I phase has a compact body-centered cubic structure, it takes a longer time for the platelets disturbed by the electric field to recover their original arrangement, as observed in Figure 1. The structural restoration process in the BP-I phase also affects the V–T curves recorded. As revealed in Figures 1 and 2, the transmittance was higher at a similar voltage on descending than on ascending. This can be explained by the body-centred cubic structure of the BP-I. As the voltage increases, the elastic force of the compact BP-I structure resists the dielectric coupling of LC molecules (helix unwinding), so that the induced birefringence is lower, giving a reduced transmittance [12]. As the external electric field in a BP system increases there will be three distinct transformations in a BP LC: (1) local reorientation of molecules caused by dielectric coupling: its response time is about 0.1 ms; (2) lattice distortion in the BP-I with a response time about 10 ms; and (3) transition to a phase of lower symmetry (e.g., transformation from BP-I to a BP-II structure), with a response time of a few seconds or more [13].

We have analysed our observations by analogy with the simulation results previously reported by Tiribocchi *et al.* [14]. According to their simulated

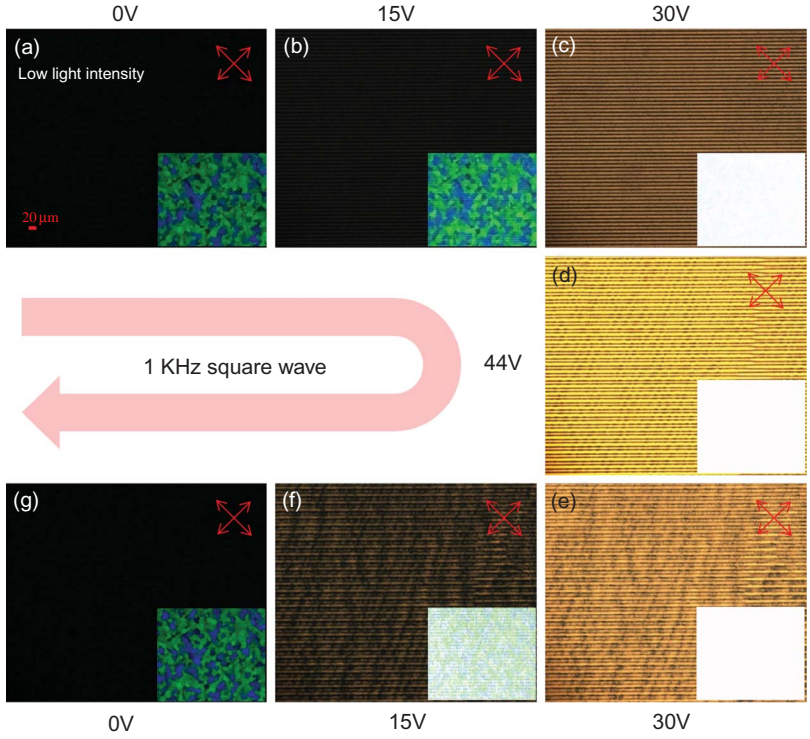

Figure 1. Textural observation of the BP-I state after applying a 1 kHz square-wave. When V_{rms} was increased to 44 V the cell reached the brightest state (d). When the field was decreasing, a multi-domain platelet texture of the BP-I was observed with significantly different brightness (b)–(f) and (c)–(e), indicating high hysteresis. In addition, the initial and final textures after one cycle of applied voltage were found to be slightly different. For clarity, textures taken with two different light intensities are shown, the main frames and insets corresponding to normal and very strong backlight intensity, respectively.

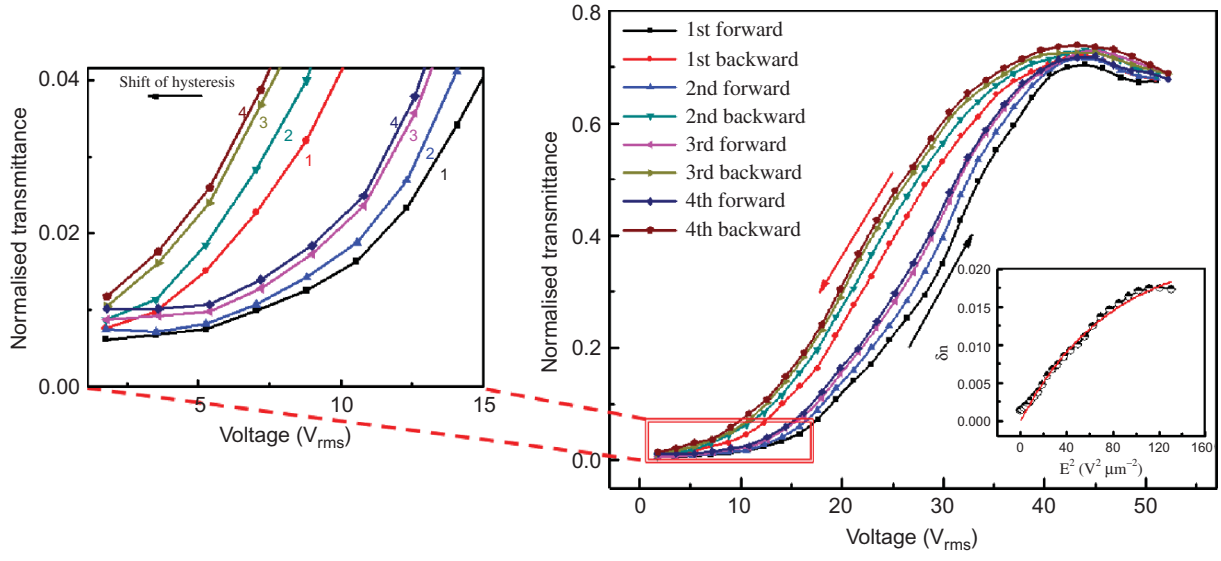

Figure 2. Measured V–T hysteresis curves of the IPS cell in the BP-I state (62°C) over four ascending and descending voltage cycles. Shift in hysteresis is shown on a larger scale. Inset shows measured refractive index change with the square of the electric field with the extended Kerr effect (solid line).

figures, when switching on, the BP-I disclinations rapidly become twisted. When the twisted defects become close enough they recombine and are once again joined at the boundaries. After the field is switched off different disclinations rejoin and pursue different dynamics, some migrating back to the centre

while others rejoin neighbouring disclinations to form either arcs connected to the same surface or lines spanning the whole device. In this case the network then becomes trapped in a metastable condition, distinct from either the zero or the high field BP-I configurations. The entire process gives rise to a hysteresis effect in the BP-I. This metastable state is quite different from the initial state and this is confirmed from the minute difference observed in Figure 1(a) and 1(f), which are dissimilar in nature. According to earlier studies, the free energies at the beginning and end of the on–off cycle are quite different, and the final state usually has a higher degree of free energy than the initial state, which is the reason for the metastability of the final state.

We observed a shift in hysteresis cycle towards the lower voltage side in successive cycles, as presented in Figure 2. This may be attributed to the fact that the initial state for the first and second cycle is different. For the BP-I phase, the initial state of second cycle has a higher value of free energy compared to that of the first cycle, due to its metastable nature [14]. After a few cycles the shift in hysteresis is not significant, which is also evident from Figure 2. The electro-optical properties of the BP can readily be explained by the Kerr effect [15], expressed as:

$$\delta n = \lambda K E^2 \tag{1}$$

where δn, λ and E are the induced birefringence, the wavelength and the strength of the electric field, respectively. K is the Kerr constant, which is a dominant material parameter indicating the magnitude of the Kerr effect. Equation (1) is valid only in the low electric field region; otherwise, it leads to a divergence as E increases. To determine the saturation trend of the experimental data, the following exponential convergence model proposed by Yan *et al*. [16], known as the extended Kerr effect, has been applied to derive the value of the Kerr constant as shown in the inset of Figure 2:

$$\delta n = \delta n_{\text{sat}} \left(1 - \exp\left[-\left(\frac{E}{E_{\text{s}}}\right)^2\right]\right) \tag{2}$$

where δn_{sat} is the saturated ordinary refractive index change and E_{s} represents the saturation field. Since we used a white light source in our experiments we have taken $\lambda = 550$ nm. In our case the value of K is found to be 1.5 nm V^{-2} and the rise in measured response time with a 1 kHz square wave for this multi-domain BP state at 62°C is found to be 4.4 ms.

When the sample was driven with a 60 Hz square wave a novel optically isotropic LC phase was observed which was completely distinct from the initial BP phase. The process of obtaining the optically isotropic state is illustrated in Figure 3. On increasing the 60 Hz square wave voltage to 80 V, the initial platelet texture (Figure 3(a)) became a featureless white state (Figure 3(b)). If the external field was decreased to 0 V, a completely dark state was induced (Figure 3(c)), which remained for only a few seconds, and was slowly transformed to the platelet texture, which was quite distinct from the initial BP-I texture. As seen in Figure 3(*c*), the sample texture was quite uniform, without any domains being visible. The light transmittance of this transient dark state showed no angle dependence, indicating an optical isotropy of the phase throughout the visible wavelength range.

The emergence of the optically isotropic state at other low frequencies was also examined, e.g. 10 Hz in Figure 3(d)–3(f). In this case, with low light intensity in the on state , some stripes occurred in the texture due to electro-hydrodynamic instability, and at 0 V the optically isotropic state was more stable than at 60 Hz .

We also checked the hysteresis behaviour of the newly observed optically isotropic state. As shown in Figure 4, switching occurred between entirely different states. In this case the optical images exhibited uniform grey levels at different applied voltages without showing any platelet-like domains. It had the appearance of a uniform mono-domain throughout the active areas – at least at the experimental optical resolution – and the optical images for ascending and descending voltage displayed no noticeable discrepancy in optical transmission. It was therefore evident that this transient state was an optically isotropic LC phase similar to BP III, although it is at present unclear what its director configuration is and why it is optically isotropic at zero field.

In the texture of the optically isotropic phase, neither platelet textures featuring BPI and BPII phases nor oily streak textures indicating the characteristic of the chiral phase were observed, making the origin of the phase unclear. A number of different electric field effects have been reported on BP samples, including field-induced transitions and distortions of the cubic lattices that form the mesophase structure. According to earlier reports, on application of electric fields to BP samples a phase transition to a turbulent chiral nematic phase is possible, and this transition depends on the frequency and magnitude of the applied electric field [13]. In addition, distortion of the blue phase in a low electric field has been demonstrated earlier in a number of studies as a shift in the Bragg reflection to longer wavelengths [17]. In the case of a 60 Hz square wave the process of disclination joining after

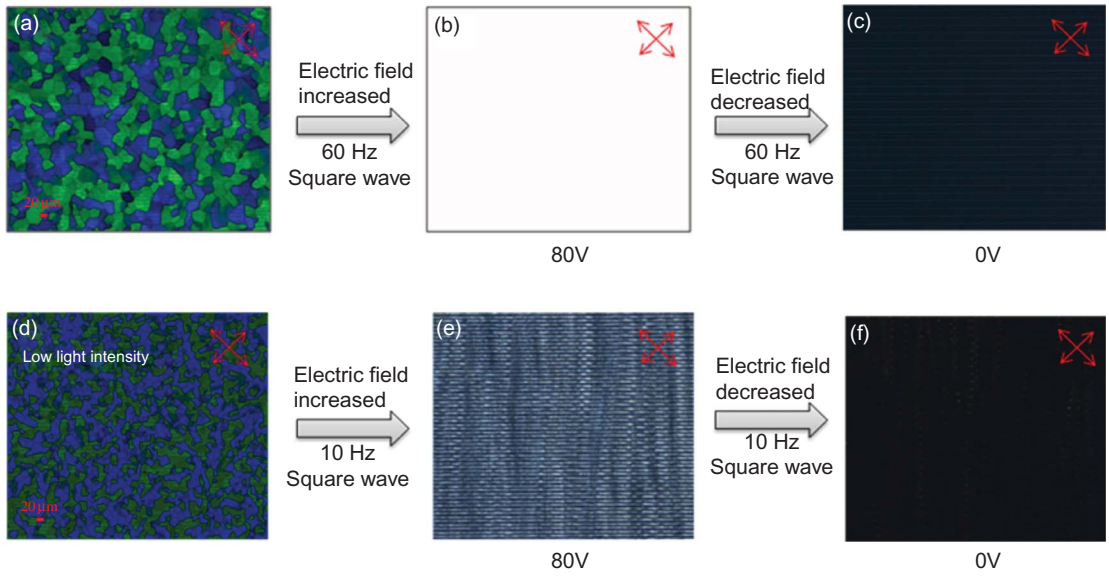

Figure 3. Emergence of the novel optically isotropic phase after driving the IPS cell with a low frequency square wave. When the cell was driven by a 60 Hz square wave the multidomain texture (a) transformed to a bright state (b), after decreasing the field at 0 V the optically isotropic state was achieved. Similar features with a 10 Hz square wave and low light intensity are shown in (d)–(f).

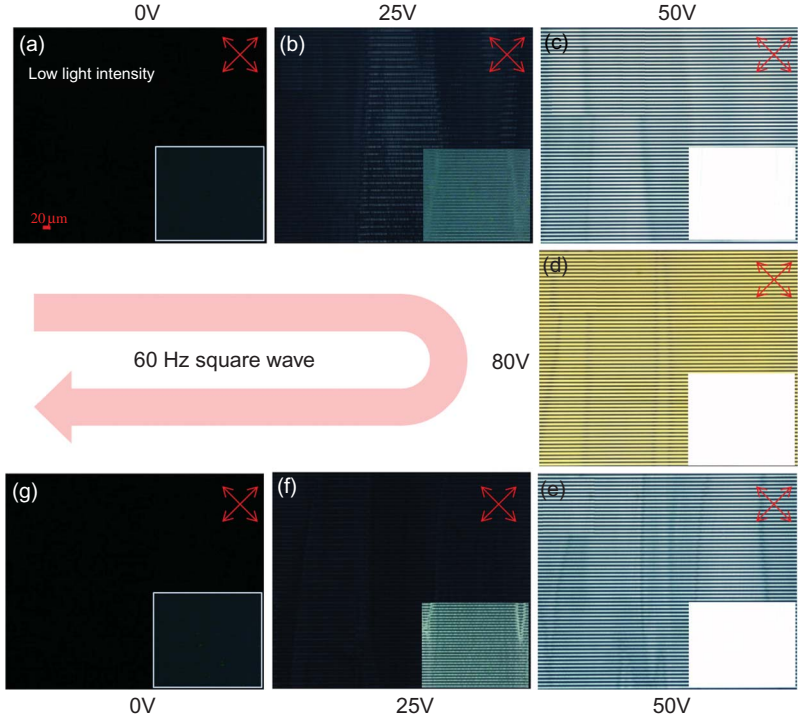

Figure 4. Textural observation of the optically isotropic state after applying a 60 Hz square wave. When V_{rms} was increased to 80 V, the cell reached the brightest state (d). While the field was decreasing, a similar type of texture was observed with almost same brightness (b)–(f) and (c)–(e), indicating low hysteresis. For clarity, textures taken with two different light intensities are shown, the main frames and insets corresponding to normal and very strong backlight intensity, respectively.

removal of the field may be slower owing to the vigorous movement of ions in the system, arising from the application of the low frequency square wave. Ionic impurities are always present in liquid crystal devices due to molecular degradation or contamination during the fabrication processes [18, 19]. Indeed, ions can

also be incorporated into the LC during sample cell preparation or by contamination of the substrates. When a low frequency AC voltage is applied to the LC cell the ions tend to be strongly separated due to modification of the time and space dependence of the electric field inside the cell. When a 60 Hz square wave is applied to the sample, and the voltage applied is high enough, all the molecules become aligned along the direction of the electric field. As the amplitude of the square wave decreases, however, the to and fro motion of ions between the electrodes limits the ability of the molecules to return to the initial double-twisted cylindrical BP-I phase. The molecular frustration of short-pitch cholesterics may also enhance this effect, and as a result the LC molecules may form very small randomly oriented LC domains. This might be the origin of the optically isotropic phase.

We have also observed a similar optically isotropic phase by applying a low frequency square wave from other LC mixtures (results not shown). In this case the limit of the low frequency applied to achieve the state was different. When the electric field was turned off the platelet-type multi-domain BP-I texture recovered after a few minutes, but the texture was found to be different to the original texture. In addition to the ideal dark state originating from the optically isotropic state, the switching behaviour offers a crucial advantage in practical applications.

Interestingly, this state shows a similar magnitude in the electro-optical Kerr effect to that in the BP-I phase. If the frequency is kept low, close to 60 Hz, essentially the same S-shaped V–T curve is observed on varying the amplitude of applied voltage as in the multi-domain BP-I state, as shown in the inset (a) of Figure 5. However, when the field was decreased, the multi-domain BP-I state became transformed into an optically isotropic state. The relative intensity of this new state was little lower than that of initial platelet type BP-I state. As demonstrated in Figure 5, the hysteresis of the new optically isotropic state with a 60 Hz square wave is noticeably reduced compared to the initial BP-I state, which corroborates the optical observations shown in Figure 4. The small difference in the transmittance level between the ascending and descending voltage cycle lies within the error limit of i-solution software. We can expect this optically isotropic phase to be a metastable state.

Using the same extended Kerr model [16] described earlier we measured the Kerr constant of this optically isotropic state, also as shown in the inset (b) of Figure 5. The measured K-value of the optically isotropic LC phase was about 1.5 nm V^{-2}, which was similar to the value obtained for the initial multi-domain BP-I phase. The operating voltage for this

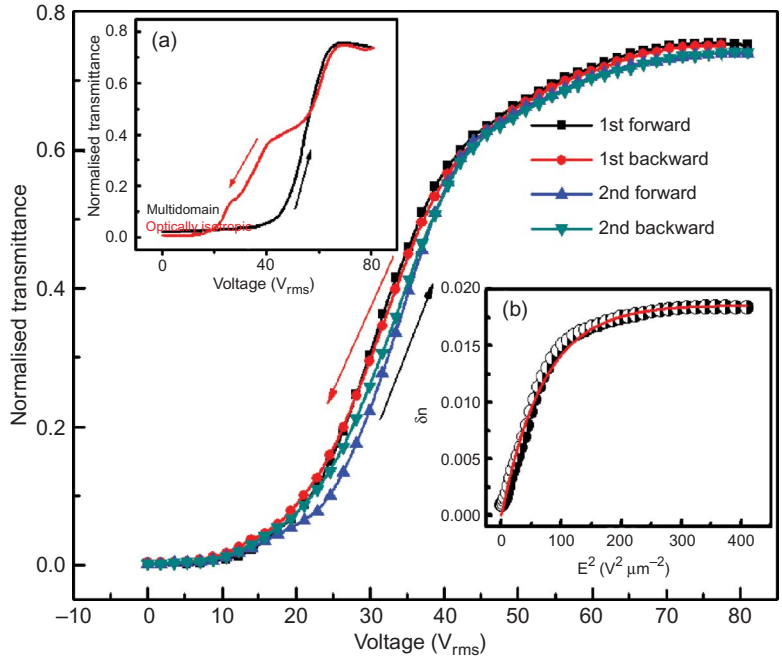

Figure 5. Measured V–T hysteresis curves of the achieved optically isotropic state of the sample driven by a low frequency (60 Hz) field. Inset (a) shows the V–T hysteresis when the multi-domain BP-I state was driven by a low frequency (60 Hz) square wave. Inset (b) shows the measured refractive index change with the square of the electric field with the extended Kerr effect (solid line) for the optically isotropic state.

phase was found to be slightly higher than for the initial BP-I state. This phenomenon can be attributed to the voltage screening effect of counter-ions. The ionic impurities can follow the low frequency square wave and hence at a particular moment a low electric field is induced by the charge separation of ions, and this is opposite to the applied electric field. Thus the electric field inside the sample is very different from that applied externally. Consequently, the transmittance in this state becomes saturated at a higher applied voltage.

The newly observed optically isotropic state shows no electro-optical hysteresis . This might be due to the lower intermolecular forces in this state compared to BP-I. The distortion of molecular arrangements is very small, which is clear from the small degree of birefringence induced on application of the electric field. As a result, a hysteresis-free behaviour is observed with this interesting optically isotropic state. Further studies are required to confirm the origin of this switchable phase, as well as its hysteresis-free nature.

We also measured the response time for this optically isotropic phase ($\sim$ 2 ms), which was lower than that of the multi-domain BP-I phase at a similar temperature.

4. Conclusions

In this study we have investigated the frequency-dependent effects of electro-optical hysteresis in the cholesteric BP-I state. We have demonstrated completely distinct switching routes, determined by the frequency of the electric field applied, and the electro-optical characteristics of BP-I are presented. The large hysteresis of the conventional BP-I phase are explained on the basis of the simulation model given by Tiribocchi *et al*. When the cell was driven by a low-frequency square wave a new hysteresis-free optically isotropic state exhibiting a high Kerr constant has been achieved. The ideal dark and grey levels with little hysteresis of the low frequency cycle has high potential in practical display applications. Further studies to identify the structure of the new state and its behaviour under an electric field will be undertaken.

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