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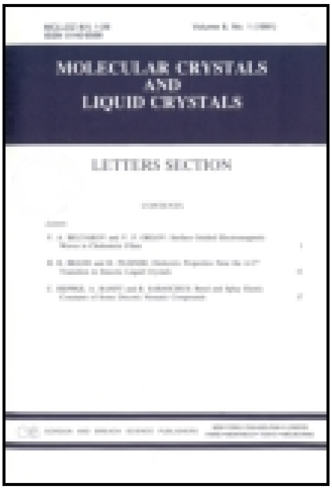

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Molecular Design of Blue Phase Materials for Display Devices

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Molecular Design of Blue Phase Materials for Display Devices

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We report that coupling of molecular biaxiality and chirality is effective for blue phase stabilization. We show a U-shaped binaphthyl derivative with a cubic blue phase and some T-shaped compounds with an amorphous blue phase III (BPIII). We designed a ternary system consisting of a conventional nematic mixture, a T-shaped BPIII stabilizer, and a chiral dopant. BPIII of the system exhibited high transmittance, submillisecond response, and hysteresis-free switching at room temperature. Amorphous BPIII, with a level of symmetry equal to that of an isotropic liquid, is thought to have a superior potential for use in next-generation displays.

Keywords Liquid crystal; blue phase; electro-optical effect; display; chirality

1. Introduction

Liquid crystals are characterized by birefringence because they have optical anisotropy. However, some liquid crystals, such as cubic and blue phases, exhibit optical isotropy. Blue phases are of particular interest because they have a fluid lattice for which the structure is stabilized by lattice defects. Believed to consist of double-twist cylinders, they are classified into three categories depending on the cylinders' packing structure: blue phase I (BPI), blue phase II (BPII), and blue phase III (BPIII) [1]. The packing structure of BPI is a body-centered cubic structure; BPII has a simple cubic structure [2, 3]. BPI and BPII are highly ordered: their disclination networks form a regular cubic lattice. In contrast, the BPIII structure has remained unresolved. Theorists have proposed that BPIII consists of double-twisted cylinders with arbitrary orientation [4, 5]. Recently, Henrich et al. proposed that BPIII is an amorphous network of disclination lines. That network is thermodynamically and kinetically stabilized over crystalline blue phases at intermediate chiralities [6]. Blue phases are recognized to offer the potential for applications as fast light modulators or tunable photonic crystals, but their narrow temperature range is a daunting obstacle to their application. Kikuchi et al. reported polymer-stabilized blue phases for which the temperature range is extended to more than 60 K [7]. Fast electro-optical switching with a response time of 10^{-4} s was demonstrated in the polymer-stabilized BPI. A blue-phase liquid crystal display (LCD) mode using polymer-stabilized BPI was developed in earlier studies [8–12]. Blue phase liquid crystals are emerging as strong contenders for use in

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*Address correspondence to Atsushi Yoshizawa, Department of Frontier Materials Chemistry, Graduate School of Science and Technology, Hirosaki University, 3 Bunkyo-cho, Hirosaki, Aomori, 036-8561, Japan. E-mail: ayoshiza@cc.hirosaki-u.ac.jp next-generation display technology. In comparison to conventional nematic LCDs, the blue phase LCD presents several important benefits: 1) it requires no alignment layer; 2) its response time is in the submillisecond range, which enables color-sequential displays; 3) its dark state is optically isotropic so that its viewing angle is wide and symmetric without optically compensating filters; and 4) it has cell gap insensitivity [13]. However, high operating voltage, hysteresis, and residual transmittance have remained. Many efforts to improve these problems have been performed from device and materials [14, 15].

We report here molecular design of blue phase materials for display devices and show hysteresis-free electro-optical switching in amorphous BPIII consisting of low-molecular-mass liquid crystals.

2. Molecular Design of Blue Phase Materials

Usually, blue phases are found in an extremely narrow temperature range (less than 2 K) between the isotropic liquid and the chiral nematic (N*) phase of sufficiently short pitch. Therefore stabilizing the blue phases has attracted much attention [16, 17]. The most successful approach is polymer stabilization [7]. Specific polymer networks can stabilize the lattice defects of a cubic BP. Blue phases are also stabilized by mixing surface-functionalized nanoparticles with chiral liquid crystals. The nanoparticles aggregation at disclination lines is responsible for BP stabilization [18]. Apart from stabilizing the lattice defects, the molecular design of blue phase materials has been investigated. Chirality is the most important factor responsible for the appearance of blue phases. Elasticity is also recognized to affect blue phase stabilization. Doping an achiral bent-core molecule is known to show marked enhancement of the BP range [19, 20]. Choi et al. reported that the stability of BPI is enhanced when K_{33} is smaller than K_{11} [21]. This effect is interpreted by the reduction in the free energy cost for defects, stabilizing the double twist cylinders. According to Fukuda [22], a blue phase temperature range can be described as equation (1),

$$\begin{aligned} &\Delta T = \frac{\tilde{K}}{8a\,R_{\max}^2}\exp\left(\frac{2K_{11}+0.57K_{22}-0.50K_{33}}{\tilde{K}} - 1\right) \\ &\text{where} \\ &\tilde{K} = \frac{1}{2}\left(K_{11}+K_{33}\right)\left[1-\frac{25}{72}\left(\frac{K_{11}-K_{33}}{K_{11}+K_{33}}\right)^2\right]. \end{aligned} \tag{1}$$

Coefficient a can be estimated from the latent heat of the chiral nematic–isotropic phase transition. Rmax is the radius of the double twist cylinders. Fukuda demonstrated that 1) the stability of BPs is greatly enhanced when the bend elastic constant K_{33} is smaller and 2) larger splay (K_{11}) and twist (K_{22}) elastic constants also stabilize BPs. $R_{\max}$ is proportional to a helical pitch. Therefore large helical twisting power (HTP) also contributes to the BP stability. However, we reported that there is no clear relation between HTP value and BP temperature range for some binaphthyl derivatives [23, 24]. Recently, Takezoe et al. noted that: 1) chiral dopant with large HTP values can easily induce BPs only by small dopant ratio and 2) HTP values were found not to affect BP stabilization directly, i.e., a wide temperature range of BPs can be obtained in mixtures containing chiral dopants with relatively small HTP values [25]. Furthermore, Alexander and Yeomans stated, based on their numerical results, a striking increase in the stability of BPI in systems where the chiral nematic phase undergoes helical sense inversion [26, 27]. Chiral effects on the BP stability are not so simple as described in equation (1).

We surmise that a molecule producing more than two helices instead of a single helix can stabilize the formation of double-twist structures. Generally a rod-like molecule

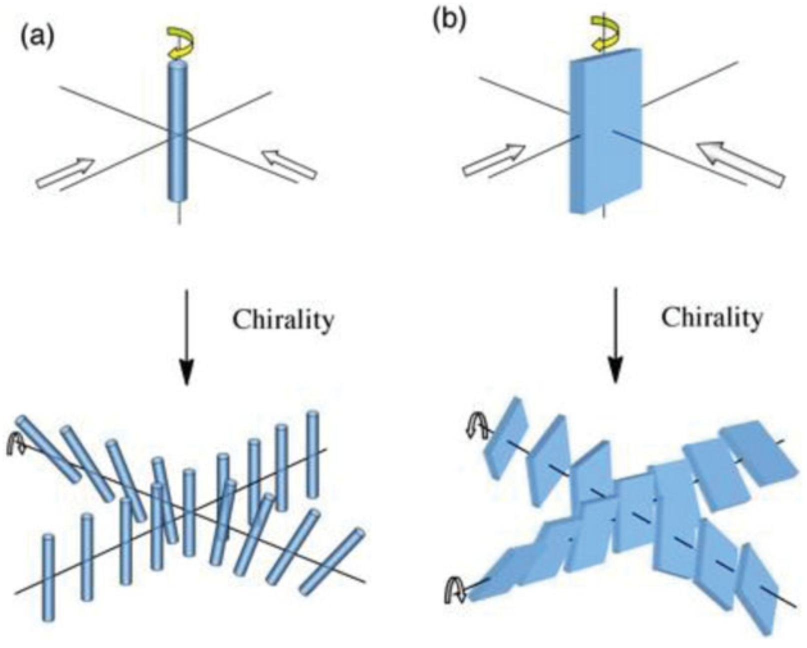


Figure 1. Helical structures of (a) a rod-like molecule and (b) a board molecule [28].

rotates freely around its long axis. Therefore all short axes perpendicular to the long axis are identical. A rod-like molecule has one distinguishable short axis. With introduction of chirality to a nematic rod-like molecule, the helical axis forms along the short axis in the chiral nematic phase [Fig. 1(a)]. By contrast, a board molecule might have two distinguishable axes. Coupling between chirality and the two short axes might induce biaxial helices that can stabilize double-twist structures [Fig. 1(b)].

We prepared a homologous series of catechol derivatives (**I-*n***) and one of biphenyl derivatives (**II-*n***) as presented in Fig. 2 [28]. We investigated the phase transition behaviour of each U-shaped compound doped with 10 wt% of a chiral dopant **ISO-(6OBA)$_2$**. The

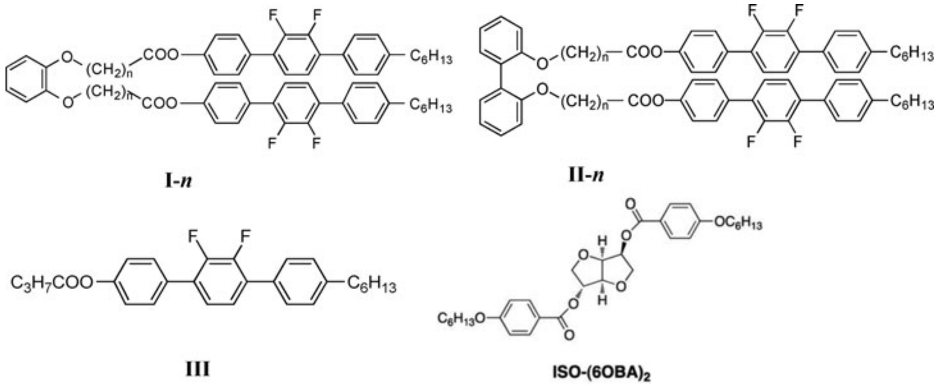


Figure 2. Molecular structures of U-shaped compounds **I-*n*** and **II-*n***, the corresponding monomeric compound **III**, and chiral dopant **ISO-(6OBA)$_2$** [28].

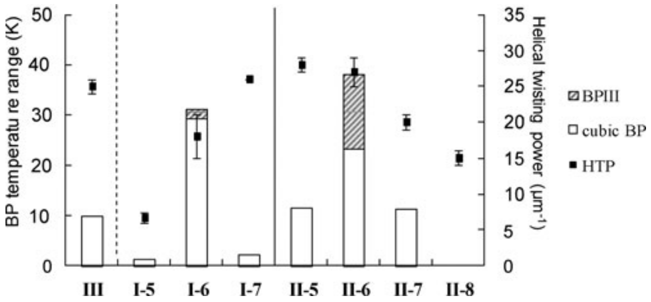


Figure 3. Relation of helical twisting power and the BP temperature range for **I-*n***, **II-*n***, and **III** [28].

appearance of the blue phases was discussed in terms of helical twisting power, elasticity, and molecular biaxiality.

Figure 3 shows the relation of helical twisting power and the BP temperature range for **I-*n***, **II-*n***, and **III**. Helical twisting power for the **I-*n*** series increases continuously with increasing spacer length, whereas that for **II-*n*** series decreases continuously with increasing spacer length. The stability of blue phases for the U-shaped system cannot be explained in terms of the helical twisting power. We observed splay (K_{11}) and bend (K_{33}) elastic constants in a nematic phase of a host nematic liquid crystal (JNC-0001) doped with 10 wt% of each U-shaped compound. Both **I-*6*** and **II-*6*** induced a marked decrease in K_{33}. It can be said that decreasing K_{33} stabilizes the BP. Rigid board molecules might have two short axes, as presented in Fig. 1(b). Such a molecule is thought to have two molecular planes that are mutually perpendicular. Each plane has a length (L)/ breadth (D) ratio. $(L/D)_{\mathrm{large}}/(L/D)_{\mathrm{small}}$ is used as a parameter describing the molecular biaxiality. This value is therefore denoted as a biaxial parameter. Figure 4 presents a relation of biaxial parameter and BP temperature range for the **I-*n*** and **II-*n*** series. An interesting finding is that a U-shaped compound possessing the largest biaxial parameter induces the widest BP range in the chiral mixture among each series, which supports the hypothesis that coupling between chirality and molecular biaxiality stabilizes the double-twist structures of BPs.

A chiral U-shaped binaphthyl derivative as shown in Fig. 5 exhibited a phase sequence of iso liq–blue phase–smectic phase on cooling [29]. Temperatures of the blue phase with a cubic structure were about 30 K. Furthermore, temperature-dependent helical twist inversion was found to occur for a mixture of the compound (50 mol%) and the nematic LC, 4-(4-hexylphenyl)-1-(4-propyloxyphenyl)-2,3-difluorobenzene, (50 mol%) in the chiral nematic phase [24]. This helical twist inversion suggests that the compound might have

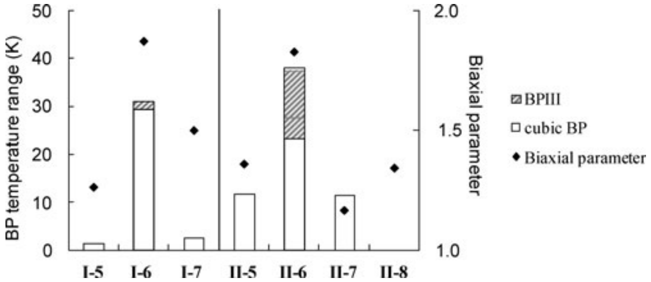


Figure 4. Relation of the biaxial parameter and BP temperature range for **I-*n*** and **II-*n*** series [28].

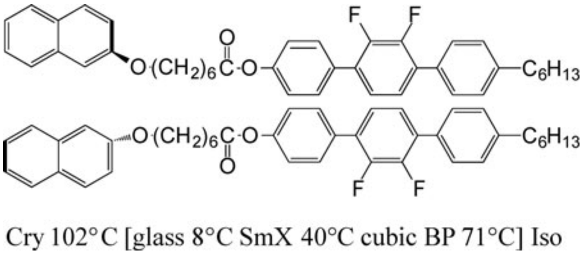


Figure 5. Binaphthyl derivative exhibiting a cubic blue phase [29].

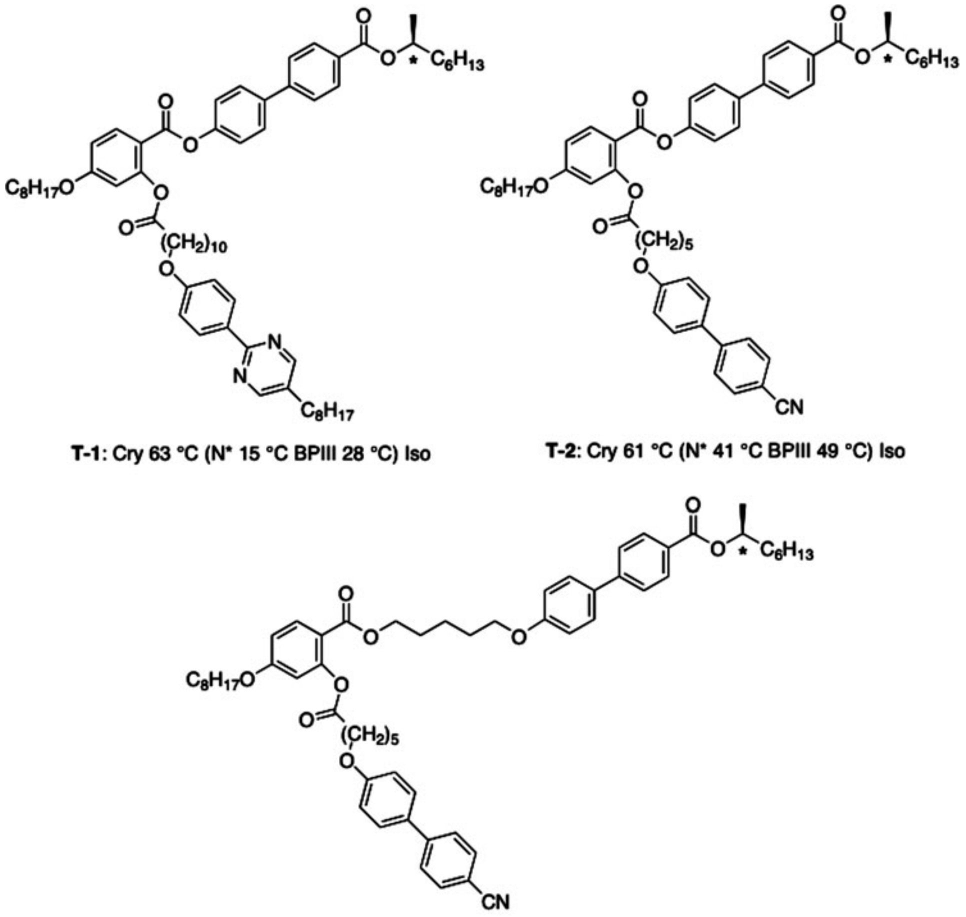
two origins of the helical twisting power. A similar relation between the appearance of BP and helical twist inversion was observed for cyclosiloxane-based side-chain co-polymer liquid crystals [30].

The structure–property relation of the U-shaped systems reveals the following design concepts of blue phase materials: 1) chirality is necessary to induce a blue phase, however, high twisting power is not prerequisite for widening the BP temperature range, 2) decreasing K_{33} with respect to K_{11} widens the BP temperature range, and 3) molecular biaxaility plays an important in producing a double-twist structure.

The U-shaped compounds prefer to stabilize a cubic BP than an amorphous BPIII. To stabilize BPIII, we designed some chiral T-shaped compounds that are also thought to have molecular biaxiality. Figure 6 shows their molecular structures. Compound **T-1** exhibited BPIII with a temperature range of 13 K on cooling [31]. Then we introduced a polar group into the chiral T-shaped system to couple the molecule with an electric field. Consequently, prepared compound **T-2** possessing a terminal cyano group exhibited BPIII and N* phases on cooling [32]. Electro-optical switching was observed in the BPIII. Recently, a dielectric study of **T-2** in the Iso and BPIII suggests that two different modes of rotation around the short axis can exist in BPIII, supporting the inference that the molecular biaxiality affects the BP stability [33]. Compound **T-3** with a flexible spacer was found to expand the BPIII temperature range up to 25 K including room temperature, on cooling [34]. Furthermore, the BPIII changed to a glass phase.

3. Comparison of Electro-optical Effects between Amorphous BPIII and Cubic BP

A mixture of **ISO-(6OBA)$_2$** (10 wt%) and compound **II-*6*** (90 wt%) exhibited both amorphous BPIII and cubic BP [29]. The phase transition temperatures were the following: Iso 85.4°C BPIII 70.6°C cubic BP 47.3°C N* 37.3°C SmX. The temperature ranges of the BPIII and the cubic BP were, respectively, 14.8 K and 23.3 K. Therefore we comparied electro-optical properties in amorphous BPIII with those in cubic BP using the single mixture [29]. Optical transmittance, as a function of an applied electric AC field at 10 Hz, was observed for a sample contained in the region between comb-type interdigitated electrodes under crossed polarizers. The transmittance in the BPIII at 80°C without an electric field was 0%. The transmittance increased concomitantly with the increase of the electric field. With an applied field of 9 V·μm^{-1}, the texture showed a homogeneous bright state with transmittance of 80%. The transmittance decreased with the decrease of the electric field. The backward curve was as the same as the forward curve. It is free from both hysteresis and residual transmittance. The absence of a lattice structure in the BPIII produces



T-1: Cry 63 °C (N* 15 °C BPIII 28 °C) Iso

T-2: Cry 61 °C (N* 41 °C BPIII 49 °C) Iso

T-3: Cry 75 °C (glass -8.2 °C BPIII 24.7 °C) Iso

Figure 6. Molecular structures and phase transition temperatures of chiral T-shaped compounds.

the hysteresis-free switching. In the cubic BP at 60°C, transmittance without an electric field was 4.6%. Although the transmittance increased concomitantly with the increase of the electric field, it did not reach a saturated value with an applied field of 12 $\mathrm{V}\cdot\mu\mathrm{m}^{-1}$. Threshold and saturated voltages in the cubic BP were higher than those in the BPIII.

We were not able to observe the backward process because the transmittance under an AC field showed time-dependent decay. Such decay was not observed in BPIII. Time-dependent transmittances under the AC field of 12 $\mathrm{V}\cdot\mu\mathrm{m}^{-1}$ are portrayed in Fig. 7. The marked difference in the electro-optical response between BPIII and cubic BP is attributed to the difference in the blue phase structure between them. Amorphous BPIII without three-dimensional order is more stable against an electric field than cubic BP. The response time in the BPIII was almost same to that in the cubic BP. At the present system, the response speed is very slow because of the small dielectric anisotropy of compound **II-*6***. Comparing the voltage-dependent transmittance in the BPIII and the cubic BP of a single mixture, the BPIII was found to have the following advantages: 1) free from hysteresis, 2)

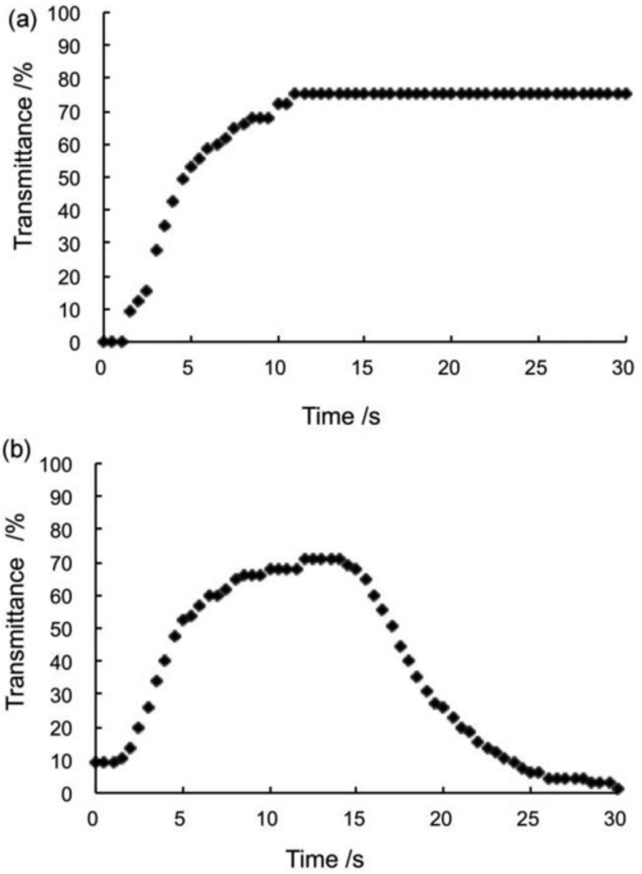


Figure 7. Optical transmittances of the mixture **ISO-(6OBA)2** (10 wt%) and compound **II-*6*** (90 wt%) as a function of time under AC field of 12 V·μm^{-1} at a frequency of 10 Hz in (a) BPIII at 80°C and (b) cubic BP at 60°C [29].

lower transmittance without an electric field, 3) lower threshold and saturated voltages, and 4) higher stability against an electric field. BPIII with the same symmetry as isotropic liquid has favourable characteristics for application to display devices: it is optically isotropic, hysteresis free, and stable in an electric field.

4. Electro-optical Switching in Amorphous BPIII

As discussed above, BPIII with the same symmetry as isotropic liquid has favorable characteristics for application to display devices. Chiral T-shaped compound T-3 exhibited BPIII with a temperature range of 25 K including room temperature upon cooling. Electro-optical switching was observed for almost all temperature ranges of BPIII [34]. However, the response times for both the rise and decay processes lengthened with decreasing temperature. The rise and decay times at room temperature are unexpectedly long. Those times at 22°C are 200 ms and 600 ms, respectively. The response times for the chiral T-shaped system are too long to use for display device applications. The long response times are attributed to large viscosity of the T-shaped compound. We designed a ternary system

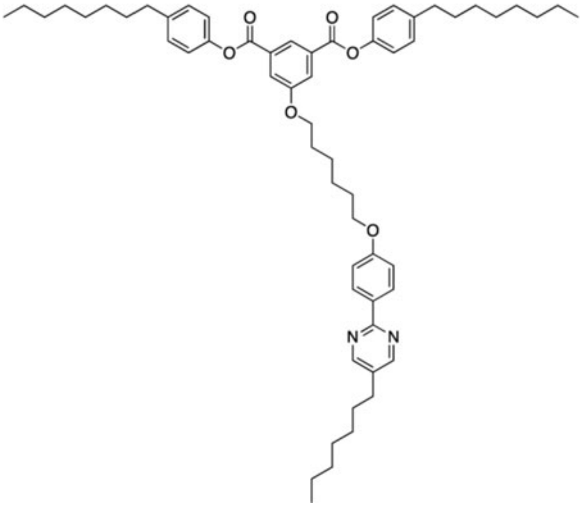


Figure 8. Molecular structure of the BP stabilizer [35].

consisting of a conventional nematic mixture, a T-shaped BP stabilizer, and a chiral dopant with high twisting power [35]. We prepared a host liquid crystal consisting of a nematic liquid-crystalline mixture **E7** (95 mol%) and a non-chiral T-shaped compound (5 mol%). The molecular structure of the T-shaped compound is portrayed in Fig. 8. **E7** consists of 4-pentyl-4′-cyanobiphenyl (51 wt%), 4-heptyl-4′-cyanobiphenyl (25 wt%), 4-octyloxy-4′-cyanobiphenyl (16 wt%), and 4-pentyl-4′-cyanoterphenyl (8 wt%). The isotropic liquid to nematic (N) transition temperature of the host LC was 52°C. The dielectric anisotropy was 12.2 in the N phase at 25°C. The host LC was doped with 15 wt% of a chiral compound **ISO-(6OBA)$_2$**. The mixture showed a phase sequence of Iso 28.4°C BPIII 21.2°C N* on cooling. The T-shaped compound exhibiting no mesogenic phase can stabilize a blue phase.

Figure 9 portrays the electric-field dependence of transmittances for the forward and backward processes in the BPIII at 26°C. The cell gap was 5 μm. The backward curve was

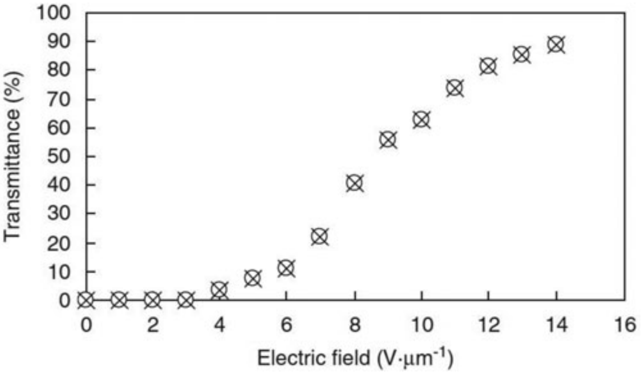


Figure 9. Optical transmittances of the chiral mixture as a function of an AC field at a frequency of 50 Hz in BPIII at 26°C. Open circles and crosses respectively denote ascending and descending processes [35].

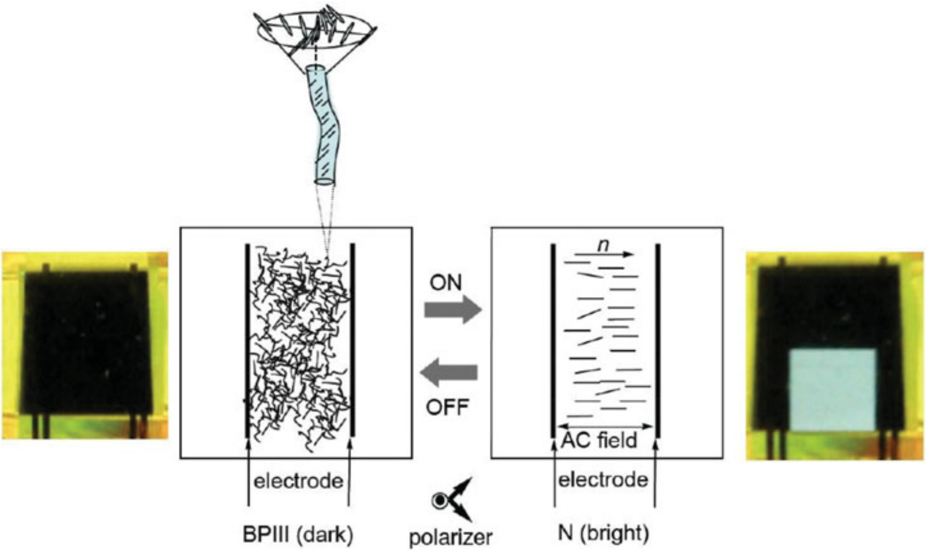


Figure 10. Schematic representation of the electric-field-induced phase transition between BPIII and N with their photographs of the evaluation BPIII cell sandwiched with cross-polarized films [35].

the same as the forward curve. It was free from both hysteresis and residual transmittance. The respective rise and decay times with an AC field of 14 V·μm^{-1} at 50 Hz were 0.4 ms and 0.8 ms. Submillisecond switching in the BPIII was demonstrated. Figure 10 presents a schematic representation of the electric-field-induced phase transition between BPIII and N with their photographs of the evaluation BPIII cell sandwiched with cross-polarized films. The BPIII cell has wide viewing angles because of its optical isotropy. After a BPIII cell had been preserved for one month at 25°C, electro-optical switching was confirmed as identical to that of the virgin state. We also presented a practical procedure by which BPIII materials are obtainable as follows: (1) develop a basic nematic mixture possessing favorable physical properties; (2) prepare a host LC by adding a small amount of a blue-phase stabilizer to the basic mixture; and (3) dope a chiral additive to the host LC.

A structure of BPIII can be stabilized by a polymer network. Chou et al. demonstrated a polymer-stabilized BPIII device with high contrast ratio and hysteresis-free switching [36]. High driving voltage and a narrow range of temperatures of the present BPIII LC are expected to be improved through appropriate material design.

5. Summary

Coupling of a biaxial molecule with a small bend elastic constant and chirality can stabilize double-twist structures, producing a blue phase with wide temperatures. Rigid U-shaped derivatives and flexible T-shaped compounds stabilize cubic BP and amorphous BPIII, respectively. We designed a ternary system consisting of a conventional nematic mixture, a T-shaped BPIII stabilizer, and a chiral dopant. BPIII of the system exhibited high transmittance, submillisecond response, and hysteresis-free switching at room temperature. Amorphous BPIII with symmetry equal to that of an isotropic liquid presents superior potential for use in next-generation displays.

Acknowledgments

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