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Fast tunable reflection in amorphous blue phase III liquid crystal

Hui-Yu Chen,a) Jia-Liang Lai, Chun-Cheng Chan, and Chih-Han Tseng
Department of Photonics, Feng Chia University, Taichung 40724, Taiwan

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The dynamic reflection spectra of amorphous blue phase III were investigated. When an electric field is applied to a blue phase III cell, the reflected wavelength does not shift obviously, but the intensity of reflection increases or decreases in a few ms. This fast intensity-tunable phenomenon in blue phase III relates to the dielectric anisotropies and chiralities of the liquid crystal and can be explained by the double twist model consisting of randomly orientated double-twisted cylinders. This study shows that blue phase III can act as a fast intensity-tunable reflector for a specific wavelength. © 2013 American Institute of Physics. [http://dx.doi.org/10.1063/1.4797492]

I. INTRODUCTION

Blue phases (BPs) appear in a narrow temperature region at the isotropic phase boundary when the chirality in the liquid-crystal (LC) material is strong enough. The difference between chiral nematic (N*) and BPs is that the LC director rotates along two orthogonal helical axes to form double-twisted cylinders (DTC) in BPs, and the rotational angle of the LC director in the DTC is from $-45^\circ$ to $45^\circ$. When these DTCs pack, due to the discontinuous orientation of the LC director around them, disclination lines form and may construct a network in the space. According to the DTC packing structure and the disclination network, BPs are classifiable into three distinct phases:1,2 blue phase I (BPI), blue phase II (BPII), and blue phase III (BPIII).1 Because the DTC packs and the disclinations form cubic lattice structures in BPI and BPII, they reflect light in a certain wavelength. The reflection spectrum of BPIII displays fog; thus it has been suggested that the structure is amorphous. Several theoretical models have been proposed to describe the structure of BPIII,3 such as an emulsion of cholesteric droplets in an isotropic matrix, the double twist model consisting of randomly orientated DTCs, the BPII-like cubic model containing a small simple cubic domain, and the quasicrystal model which is characterized by a locally icosahedral symmetry. However, to date, a suitable description of the BPIII structure has not been confirmed. Most experiments focus on studying BPI and BPII, as they are easier to observe and stabilize in a wider temperature range. Because the available temperature range of BPIII was limited to one or less Kelvin for many years, only a few papers present experimental observations of BPIII. From these previous studies, we can conclude the following experimental characteristics of BPIII: (1) BPIII is stabilized by strong chirality and usually occupies a temperature range of $\sim 0.1^\circ$C just below the isotropic phase;4 (2) BPIII is strongly optically active; (3) BPIII reflects a very weak and broad region at short wavelengths, which depends on the chirality of the liquid crystals;5 (4) the latent heat of BPIII is larger by an order of magnitude than other phase transitions6 such as N* to BPI or BPI to BPII; and (5) BPIII is only weakly affected by moderate electric fields;3,5,6 (6) because of the Kerr effect, BPIII can be switched between optical isotropy and anisotropy under an in-plane field.7,8

In our previous study,9 we found that for cubic BPI the reflection spectra can be changed by applying an external field. As the external field increases, the reflected peak shifts to a long or short wavelength, depending on the directions of the electric field and lattice surface and the dielectric anisotropy of the LC materials.5 The wavelength shift of BPI in the field can be larger than 40 nm, and the intensity of the reflection does not change obviously.9 This means that BPI can be used as a wavelength-tunable reflector.

Here, we demonstrate an intensity-tunable reflector by using amorphous BPIII materials as they are subjected to an electric field. When the direction of the electric field is perpendicular to the cell substrates, the reflection spectrum of BPIII exhibits a huge change in intensity in just a few milliseconds, and the reflected wavelength shift is less than a few nanometers. Response times, including the switching-on and switching-off times, relate to the sign of the dielectric anisotropy of the LCs. Moreover, the reflected intensity reduces (increases) as the strength of the electric field increases, when the dielectric anisotropy of the LC is positive (negative). From the relation of the reflected intensity and dielectric anisotropy of the LC samples, the change in reflected intensity in BPIII may be induced by the local director rotation constructing DTC. The dependence between the concentration of chiral dopant in the LC and the temperature range of BPIII was also tested in this study, with the results showing that BPIII can be stabilized in the weak chirality. These experimental observations offer a concept to design fast photonic devices based on BPIII materials.

II. SAMPLE PREPARATION AND EXPERIMENTAL SETUP

In order to obtain an LC material with a wider BPIII temperature range, the relation between the concentrations of chiral dopant in nematic LC and the temperature ranges of the three blue phases was studied. Chiral dopant 811 (bought from Aldrich) was added into a negative-dielectric-anisotropy nematic LC in various doping concentrations.

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a)Author to whom correspondence should be addressed. Electronic mail: huiyuchen@fcu.edu.tw.
These mixtures were filled into 9-μm cells without surface treatment. Experiments were carried out on a microscopic hot stage (Mettler) where the temperature stability is less than 0.05 °C. The temperature ranges of each blue phase were determined by measuring the reflection spectrum through a polarization optical microscope (Olympus, BX51). The UV-Vis optical spectrometer used in this study is the Ocean Optics USB4000 in which the wavelength range is detected from 350 nm to 850 nm, and the optical resolution is 0.3 nm.

Figure 1(a) is the intensity of reflected light in the BP sample when it is cooled from the isotropic phase at a rate of 0.1 °C/min. Because the reflected intensity in BPI is an order larger than that of BPIII, the transition temperature from amorphous BPIII to cubic BP is not hard to distinguish through the behavior of the reflected spectrum. The temperature ranges of BPI and BPII are determined by detecting the reflected peaks by cooling and heating the LC material. The reflected peak from BPI is strongly dependent on whether the LC material is heated into BPI from chiral nematic phase or cooled from BPI.10 The temperature-dependent reflected peak shows hysteresis in BPI, but it does not in BPII. For the mixture with 30-wt. % S811, the hysteresis of reflected peaks exhibits when the temperature of the material is below 92.5 °C, where the LC phase transited from BPIII to cubic BP. Thus, the BPII does not exist in the 30-wt. % S811 mixture. However, when the concentration of the chiral dopant is reduced to 24 wt. %, the BPII is observed between the BPI and BPIII. Figure 1(b) is the phase diagram of the three BPs when the concentration of the chiral dopant is varied. When the concentration is larger than 24 wt. %, the temperature range of BPIII is extended from 0.5 °C to 2 °C. Notice that, in our samples shown here, we found that BPIII can stabilize even with a lower concentration of chiral dopant, and its widest temperature range is around 2 °C.

### III. RESULTS AND DISCUSSION

In order to observe the dynamic reflection spectrum of BPIII under the application of an electric field, the 30-wt. % chiral dopant S811 was added into a negative-dielectric-anisotropy LC. Figure 2 is the change in the reflection spectra of BPIII by increasing the strength of the electric field. The intensity of the reflection spectra in the BPIII cell increased with the increase in the electric field, and the reflected intensity was saturated when the applied voltage was larger than 40 V. Notice that the wavelength shift of the reflection is not obvious. Moreover, the full width at half maximum reflectance (FWHM, Δλ) in BPIII reduces continuously when increasing the applied voltage. This indicates that the reflected color can be purified and enhanced when applying the electric field to the BPIII cell. Moreover, the response times for tuning the reflected intensity in the BPIII cell are less than a few ms, as shown in Fig. 3. This intensity-tunable phenomenon is not usually seen in other chiral LC systems. For cholesteric LC (ChLC) and cubic BP, the reflected wavelengths are changed by applying an electric field. At the same time, the intensity is reduced and Δλ may become wider, due to the destruction of the periodical helix structure. ChLC and cubic BP are suitable as a wavelength-tunable reflector, but they cannot tune the reflected intensity at a specified wavelength (color) as well as BPIII can, as is shown in Fig. 2.

![FIG. 1. (a) Reflected intensity versus temperature in NLC with 30-wt. % S811. (b) Phase transition temperature, relative to $T_{iso}$, as a function of chiral dopant concentration.](image1)

The influences of the dielectric anisotropy and the chirality of LCs on the reflection spectra are studied by preparing three nematic liquid crystal mixtures with different dielectric anisotropies $\Delta \varepsilon$ (LC01 $\Delta \varepsilon = 4.8$, LC02 $\Delta \varepsilon = 0.361$, and LC03 $\Delta \varepsilon = -5.1$). According to the phase diagram in Fig. 1(b), chiral dopant S811 was added into these LC hosts at 25 wt. % and 30 wt. % to obtain BPIII with a 2 °C temperature range. Comparing the behaviors of the BPIII in LC hosts
with positive and negative dielectric anisotropies, we note that, under the influence of the vertical electric field, the reflected intensity increases or reduces dramatically, and the reflected colors in each sample do not change obviously, as shown in Table I. The reflected intensity becomes stronger in the negative-dielectric-anisotropy BPIII cell (LC03) but reduces when the LC host is a positive-dielectric-anisotropy material (LC01 and LC02). A similar relation between the change in the reflected light intensity and dielectric anisotropy of LCs has also been seen in different LC material samples and in previous studies.3,6 From the spectra in BPIII in Fig. 2 and BPI in our previous study,9 we can summarize the following differences between the behaviors of BPIII and BPI (or BPII) as: (1) the maximum wavelength shift of reflectance in BPIII is less than in BPI or BPII; (2) when a vertical electric field is applied to BPIII, a dramatic change in the reflected intensity is observed; (3) the threshold field to change the reflection spectrum in BPIII is greater than in BPI or BPII; (4) FWHM $\Delta \lambda$ in BPIII is greater than it is in other BPs; (5) a phase transition from BPIII to chiral nematic does not occur even with a very strong electric field, but it does in a BPI sample; (6) the dielectric anisotropy of the LC host affects the tendency of the reflected intensity to increase or decrease in a BPII sample but does not affect cubic blue phases. These differences imply that the structure of BPIII may be different from that of other BPs with a cubic structure.

Comparing the response times of the three BPIII samples as shown in Fig. 4, one may see that the $\tau_{on}$ is slower in the BPIII with negative-dielectric anisotropy. Moreover, when the concentration of chiral dopant in the LC host is increased, these response times only slightly reduce, as can be seen in Fig. 5. The behavior of the response times in BPIII cannot be explained by the unwinding helix model in cholesteric LC, because the rise time is independent of the chirality and is affected by the sign of the dielectric anisotropy of the LC. The fast response in BPIII that we present here is associated with the reorientation of the LC molecules constructing DTCs in BPIII.

Referring a calculation of the surface- and field-induced ordering in BPIII by Kitzerow,11 the relation between the FWHM and the electric field is described as $\Delta \lambda = \Delta \lambda_0 \left[1 - \left(E/E_0\right)^n\right]^{1/2}$, where $n$ depends on the BPIII structure, and $\Delta \lambda_0$ and $E_0$ are adjustable parameters. In this study, the field-dependent FWHM data were obtained from Fig. 2, and it was fitted by $\Delta \lambda = \Delta \lambda_0 \left[1 - \left(E/E_0\right)^n\right]^{1/2}$. The fitting value of $n$ is 2.29 ± 0.5 and it is consistent with the double-twist model.11 According to this calculation and other experimental evidences, the most

TABLE I. Reflection patterns of BPIII in the vertical electric field by microscope.

<table>
<thead>
<tr>
<th>LC</th>
<th>LC01</th>
<th>LC02</th>
<th>LC03</th>
<th>LC03</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta \epsilon$</td>
<td>4.8</td>
<td>0.361</td>
<td>-5.1</td>
<td>-5.1</td>
</tr>
<tr>
<td>S811 (wt. %)</td>
<td>25</td>
<td>25</td>
<td>25</td>
<td>30</td>
</tr>
<tr>
<td>0 V/μm</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4.4 V/μm</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>11 V/μm</td>
<td></td>
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<td></td>
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</tr>
</tbody>
</table>

FIG. 3. Response in a BPIII cell with application of a 40-V external voltage (inserted pictures are the microscopic pictures of BPIII).

FIG. 4. Response times, including switching-on time $\tau_{on}$ and switching-off time $\tau_{off}$ in different BPIII samples with 25 wt. % chiral dopant. (The LC hosts are ○: LC01, ▲: LC02, and ■: LC03)
The possible structure of BPIII is that it is constructed of random DTCs and an amorphous network of disclination lines. The reflection spectrum in BPIII is weak and broad, because the helical axes perpendicular to the cylinder axes of the DTC orient randomly. After applying the electric field, the reflection peak can be sharper and stronger in the negative-dielectric anisotropy BPIII sample but weak in the positive-dielectric anisotropy BPIII sample. Computer simulation of BPIII in electric fields has been carried out and points to a field-induced transition between BPIII and BPE (field-induced BP).\(^\text{12}\) In their simulation, the phase transition to BPE can occur in samples with either positive or negative dielectric anisotropy. For negative-dielectric-anisotropy BPIII material, the electric field induces BPE, where the disclination lines lie along layers stacked parallel to the electric field.\(^\text{12}\) This means that the cylinder axes of the DTCs become ordered in space and their direction is also parallel to the electric field. By considering the structure of DTCs, the LC director rotates simultaneously on two helical axes perpendicular to the cylinder axis. When viewed along the field direction, the helical axes of the LC molecules are parallel with the scattering vector, and then reflect the sharper peak as we see in Fig. 2. Consequently, the reflected intensity increases dramatically with the electric field in the negative-dielectric anisotropy sample. The shift in reflected wavelength after applying the electric field in Fig. 2 is decided by the spacing between the disclination lines. However, for the positive-dielectric anisotropy sample, because the helical axes of the LC molecules are perpendicular to the scattering vector, the reflected intensity decreases. This indicates that the fast response in BPIII that we present here is associated with the reorientation of the LC molecules constructing DTCs in BPIII.

**IV. CONCLUSION**

In this study, we demonstrate a fast intensity-tunable reflector by amorphous BPIII LC and discuss its dynamic reflection spectra in an electric field. After applying the vertical electric field, the reflected intensity of BPIII is changed in a few ms. This change depends on the dielectric anisotropy of the LC materials. When it is positive, the reflected intensity reduces with the increase in strength of the electric field. However, in the BPIII sample with negative dielectric anisotropy, the opposite behavior is observed. From the experimental results in this study, the structure of BPIII can be explained by the double twist model, and then the change in reflected intensity is induced by the local director rotation constructing DTC. This study shows that blue phase III can be a fast intensity-tunable reflector for a specific wavelength, where the reflected wavelength is decided by the concentration of the chiral dopant and the LC host.

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