Diluter Effects on Polymer-Stabilized Blue Phase Liquid Crystals

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Abstract—Diluter effects on the phase transition temperatures, Debye relaxation frequency, and electro-optics of polymer- stabilized blue phase liquid crystal (BPLC) are investigated. Some diluters not only significantly decrease the BPLC response time but also slightly enhance the Kerr constant. For practical applications, optimal diluter concentration should be considered in order to balance the needs between response time and operation voltage.

Index Terms—Blue phase, diluter, Kerr constant, liquid crystals.

I. INTRODUCTION

P OLYMER-STABILIZED blue phase liquid crystal (PS-BPLC) [1]–[3] is a promising candidate for next-generation display [4]–[6] and photonic devices [7], [8] because of its attractive features, such as submillisecond response time [9], [10], no need for surface alignment layer, and optically isotropic dark state. However, high operation voltage still hinders its widespread applications. In addition to protruded electrode device structures [6], [11], [12], BPLC materials with a large Kerr constant (K) play a crucial role because the on-state voltage is inversely proportional to \sqrt{K} [13]. From Gerber's model [14], Kerr constant is related to the birefringence (Δn), dielectric anisotropy ($\Delta \varepsilon$), average elastic constant (k), and pitch length (p) of the LC host as

$$K \approx \Delta n \cdot \Delta \varepsilon \frac{\varepsilon_0 p^2}{\lambda k (2\pi)^2}.$$
 (1)

From (1), increasing $\Delta \varepsilon$ is a straightforward way for enhancing Kerr constant. Indeed, BPLC hosts with $\Delta \varepsilon > 100$ have been developed recently [13], [15]. For comparison, a typical nematic LC has $\Delta \varepsilon < 10$. Therefore, BPLC represents a paradigm shift in LC material development. The problems associated with such a huge $\Delta \varepsilon$ LC are threefold: 1) its viscosity is large, so that the device response time goes back to 1-ms range and the one-drop fill panel fabrication process becomes more difficult; 2) the pixel's capacitance is increased by $\sim 10 \times$, so that a longer circuit charging time is needed [16]; 3) lower relaxation frequency; e.g., JNC JC-BP01M has a relaxation frequency of 1.3 kHz, and its Kerr constant decreases by $\sim 2 \times$ as

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the frequency increases from 60 Hz to 1 kHz [17]. When driving this BPLC at 1 kHz for color sequential displays, two serious problems are encountered: 1) the operation voltage is increased by \sim 40%, and 2) the dielectric heating effect [18]–[20] due to the imaginary part (absorption) of dielectric constant, which has a peak value at the relaxation frequency, will affect the performance stability of the BPLC device.

Diluters have been widely used for lowering the viscosity and melting temperature of a nematic LC [21]–[27]. However, diluter effect on the BPLC properties has not been investigated. Here, we report the diluter effects on a BPLC host which has a large $\Delta \varepsilon$. When we dope 6 wt% of a diluter to the LC host, the mixture's viscosity (η) is dramatically reduced, while the decrease in Δn and $\Delta \varepsilon$ is mild. Most diluter molecules are fairly short, while the large $\Delta \varepsilon$ BPLCs have bulky and long molecules with several dipole groups. Thus, after the addition of diluters the average molecular length l is shortened. Since the relaxation frequency f_r is proportional to $1/(\eta l^3)$ [28], diluters help to increase f_r . This increased relaxation frequency is less critical to nematic LCs because their relaxation frequency is already high (>50 kHz), but is important to BPLCs because their relaxation frequency is usually low (<10 kHz) [14].

In this paper, several diluters, including compounds and mixtures, were employed. Doping 6% of some diluters not only reduces the response time by $2\times-3\times$, but also improves the Kerr constant. To compare the performance with and without diluters, we use a Figure of Merit (*FoM*) defined as the ratio of Kerr constant to decay time (K/τ) [29]. Higher diluter concentration helps to further decrease the response time, but its tradeoff is the decreased Kerr constant.

II. LC HOST PROPERTIES

In experiment, we used LC1 (HTG-135200, HCCH, China) as the nematic host. Its physical properties are listed as follows: $\Delta n = 0.205$ at $\lambda = 633$ nm, $\Delta \varepsilon = 99$ at 1 kHz, $\gamma_1 = 700$ mPas at 25°C, and clearing temperature $T_c = 98$ °C. We mixed 94 wt% of LC1 with 6% of a nonpolar diluter 5CC3 [4-pentyl-4'-propyl-1,1'-bi(cyclohexyl)]. For convenience, we call this mixture as LC2.

Both Δn , $\Delta \varepsilon$, viscoelastic constant (γ_1/k_{11}) and γ_1 of the LC mixture are all temperature dependent, as described by following equation [29]–[34]:

$$\Delta n = \Delta n_0 S,\tag{2}$$

 $\Delta \varepsilon = aS \exp(U/k_B T) \tag{3}$

$$\gamma_1/k_{11} = b \cdot \exp(E_a/k_B T)/S \tag{4}$$

$$\gamma_1 = cS \exp(E_a/k_B T) \tag{5}$$

LC Host	Δn_{0}	β	U (meV)	a	E _a (meV)	b	с
LC1	0.297	0.226	109	2.07	550	2.9E-8	2.3E-7
LC2	0.279	0.227	109	1.87	501	1.5E-7	7.7E-7

 TABLE I

 FITTING PARAMETERS FOR LC1 AND LC2 WITH (2)–(6)



Fig. 1. Temperature dependent (a) Δn at $\lambda = 633$ nm, (b) $\Delta \varepsilon$ at 1 kHz, (c) γ_1/k_{11} and (d) γ_1 of LC1 and LC2. Dots represent the measured data and lines are fitting results according to (2)–(5), respectively.

where S denotes the order parameter, Δn_o is the extrapolated birefringence at T = 0 K, U is a parameter related to dipole moment, E_a is the activation energy, k_B is the Boltzmann constant, and a, b, and c are proportionality constants. Order parameter S can be approximated by Haller's semi-empirical equation [35], [36]:

$$S = (1 - T/T_c)^{\beta} \tag{6}$$

where β is a material constant and T_c is the clearing point of the LC mixture. After doping 6% of 5CC3, the T_c of LC2 drops slightly from 98 °C to 96 °C.

Fig. 1 depicts the temperature dependent Δn (at $\lambda = 633$ nm), $\Delta \varepsilon$ (at 1 kHz), γ_1/k_{11} and γ_1 of LC1 and LC2. Please note that these are the LC hosts without chiral dopants and photo-curable monomers. Here, dots are experimental data and lines are fitting results with (2)–(5). Table I lists the fitting parameters. The agreement is quite good. After doping 6% 5CC3 to LC1, the Δn of LC2 is decreased by about 6%, $\Delta \varepsilon$ by 10%, and more amazingly the rotational viscosity γ_1 is reduced by 200% at 25°C [Fig. 1(d)]. However, doping a low molecular weight diluter also lowers the elastic constant. As a result, γ_1/k_{11} is only reduced by 20% [Fig. 1(c)]. From Table I, the activation energy E_a of LC2 is about 10% smaller than that of LC1 because of the lower viscosity induced by the diluter.

III. RELAXATION FREQUENCY

For color sequential displays, a higher operation frequency (up to 1 kHz) is needed in order to suppress color breakup. In a BPLC, the relaxation frequency of Kerr constant is closely related to the dielectric relaxation frequency f_r of the LC host



Fig. 2. Illustration of relaxation frequency shifting by doping a low viscosity diluter to a LC host.

[17]. For a low- viscosity nematic LC host, its f_r is usually over 50 kHz. For a BPLC host with a large $\Delta \varepsilon$, the molecules tend to be long and bulky because multiple dipole groups are employed. Therefore, its dielectric relaxation frequency is reduced significantly. For a positive $\Delta \varepsilon$ LC material, ε_{\perp} remains almost unchanged in the low frequency region due to its much higher f_r compared to $\varepsilon'_{//}$ (real part of the permittivity) [37]. The relaxation of $\Delta \varepsilon$ basically follows that of $\varepsilon'_{//}$. The frequency dependent $\varepsilon'_{//}$ was obtained by measuring the capacitance of a homeotropic LC cell (results are not shown here) and fitting the experimental data with Debye relaxation equation [38], [39]:

$$\varepsilon_{//}^{'} = \varepsilon_{\infty} + \frac{\varepsilon_0 - \varepsilon_{\infty}}{1 + (f/f_r)^2} \tag{7}$$

where ε_0 is the static permittivity along the long molecular axis at low frequency, and ε_{∞} is the permittivity at the high frequency limit. The measured f_r at 25°C for LC1 and LC2 is 12.6 kHz and 15.4 kHz, respectively. When we dope 6 wt% 5CP3 (structure shown in Table II) into LC1, f_r increases to 17.2 kHz. Diluters help to increase the relaxation frequency of the LC host due to the reduced viscosity and shortened average molecular length. This is particularly favorable for high frequency operation of the LC host and PS-BPLC. This effect becomes more critical for the LCs with low relaxation frequency, say $f_r \sim 1$ kHz. Let us illustrate this principle using following example.

As shown in Fig. 2, the LC host has a very large $\varepsilon'_{//}$ (190; black solid line) at 100 Hz, but it declines sharply as the frequency increases. At $f_r = 500$ Hz, $\varepsilon'_{//}$ drops by 2X, and at 1 kHz it further decreases to 40. By adding a diluter, the relaxation frequency shifts rightward to 2 kHz (red solid line). Although at 100 Hz, its real part is decreased from 190 to 160, at 1 kHz (the intended operation frequency for color sequential displays) its $\varepsilon'_{//}$ remains as large as 130 (cf., 40 for the host

PSBP #	LC host	Diluter type	Diluter Structure	<i>K</i> (nm/V ²)	Decay time (τ, μs)	FoM* (K/τ)	<i>T_{m_BP}</i> (°C)	<i>T_{c_BP}</i> (°C)
1	LC1	No diluter		8.27	1700	1	5.6	82.3
2	LC2	5CC3	H ₁₁ C ₅	8.04	573	2.90	7.9	81
3	LC3	5CP3	H ₁₁ C ₅ -C ₃ H ₇	8.83	740	2.46	-1	75.4
4	LC4	5CCV	H ₁₁ C ₅ -	7.48	1200.8	1.29	-4.5	79.5
5	LC5	5CP02	H ₁₁ C ₅	9.11	988	1.90	0	81
6	LC6	5PP3	H ₁₁ C ₅	9.44	737.3	2.64	4.5	76
7	LC7	4CC3		7.39	633.4	2.41	3	80.5
8	LC8	5CC2		7.4	678.7	2.25	1	79.3
9	LC9	5CAP02	H ₁₁ C ₅	7.36	738.2	2.06	-1.1	78.2
10	LC10	MLC-6686	5	7.36	1062.4	1.43	3.5	81.5
11	LC11	FPPF	FF	6.37	638.4	2.06	-13	72.5
12	LC12	5CC3 & 40	CC3 at 1:1 ratio	7.44	792	1.94	4.2	81.5

TABLE IIPHYSICAL PROPERTIES OF PSBP SAMPLES CONTAINING 6% OF DIFFERENT DILUTERS. HERE, KERR CONSTANT AND DECAY TIME WERE MEASURED AT $\lambda = 633$ NM and 25°C. (*the FoM is Normalized to that of PSBP-1)

without diluter). Meanwhile, the dielectric absorption (imaginary part $\varepsilon_{//}^{"}$) peak also shifts to a higher frequency (red dashed lines), which helps to reduce dielectric heating effect.

IV. DILUTER EFFECT ON PS-BPLC

To investigate the diluter effect on a PS-BPLC without affecting its pitch length and monomer concentration, we keep the ratio of LC host to chiral dopant/monomer the same for the 12 PSBP samples studied. In Table II, the eleven LC hosts (LC2-12) all consist of 94 wt% of LC1 and 6 wt% of different diluters. We prepared BPLC mixtures with 4.5 wt% chiral dopant R5011 (HCCH) and 95.5 wt% LC host. Afterwards, 8 wt% of photocurable monomers (4.8 wt% RM257 (Merck)+3.2 wt% C12A, Sigma Aldrich) and 0.5 wt% of photoinitiator were blended with 91.5 wt% of the BPLC mixture to form the precursor. Next, we injected the LC/monomers mixture into an in-plane-switching (IPS) cell in an isotropic phase. The IPS electrode width and gap are 10 μ m and the cell gap is $\sim 7.3 \ \mu m$. The cell was placed on a Linkam heating/freezing stage controlled by a temperature programmer (Linkam TMS94) and cooled to BP phase and then exposed to UV light ($\lambda = 365$ nm) with intensity of 8 mW/cm² for 10 min. After UV irradiation, the BPLC nano-composites were self-assembled and the blue phase to isotropic phase transition temperature (T_{c_BP}) for PSBP-1 was 82.3 °C. With diluters, the T_{c_BP} is few degrees lower, as listed in Table II, because of the relatively low clearing point of the diluter itself. The impact of 1°C–2 °C decrease in $T_{c_{-BP}}$ with some diluters is not significant. On the other hand, the chiral nematic to blue phase transition temperature (T_{m_BP}) could be lowered by adding a proper diluter, which helps to increase the low temperature stability. For PSBP-11, although 6% of FPPF lowers the T_{c_BP} by 10 °C, the T_{m_BP} is also lowered from 5.6 °C to -13 °C,

which is favorable for low temperature operation. The color of these PSPB samples looks almost the same, indicating the pitch length variation after adding diluter is negligible.

The IPS cell was mounted on the Linkam heating stage and sandwiched between two crossed polarizers. A linearly polarized He–Ne laser ($\lambda = 633$ nm) was used as probing beam. A 1-kHz square-wave AC signal was applied to the IPS cell. The light transmittance was measured by a photodiode detector (New Focus Model 2031) and recorded digitally by a LabVIEW data acquisition system (DAO, PCI 6110). Fig. 3 depicts the measured voltage-dependent transmittance (VT) curves for PSBP-1, 2, 8, and 10. The first peak occurs at 61.6 V, 62.4 V, 66 V, and 66.6 V respectively. Through fitting with extended Kerr model [4], [40], we obtained the Kerr constant $K = 8.27 \text{ nm/V}^2$ for sample PSBP-1. After adding 6% of diluters, the Kerr constant of PSBP-2, 8, and 10 decreases to 8.04, 7.40, and 7.36 nm/V², respectively. The variation is within 11%, since the effect of diluter on Δn and $\Delta \varepsilon$ is small. On the contrary, the on-state voltage of PSBP-3, 5 and 6 decreases slightly to 61 V, 61 V, and 59.6 V. This is a pleasant surprise because the employed diluters are non-polar and low birefringence compounds. We expect they would decrease the mixture's Δn and $\Delta \varepsilon$ significantly so that the on-state voltage would increase instead of decrease.

To understand this unexpected phenomenon, we have to examine what factors determine the Kerr constant. From (1), Kerr constant of a BPLC is primarily governed by the ratio of $\Delta n \cdot \Delta \varepsilon / k$ (note here k is the average elastic constant of the PSBP, which is different from the k_{11} of the LC host), provided that the pitch length and wavelength remain the same. As discussed above, after doping a diluter both Δn , $\Delta \varepsilon$, k_{11} and γ_1 decrease but at different rates. Therefore, it is possible for $\Delta n \cdot \Delta \varepsilon / k$ to either increase or decrease, depending on the relative change of



Fig. 3. Measured VT cures for PSBP-1, 2, 8, and 10. $\lambda = 633$ nm.

 Δn , $\Delta \varepsilon$, and k induced by the diluter, which is related to the sophisticated inter-molecular interactions between the elongated, bulky host LC and the short, small-sized diluter molecules.

Fast response time is one of the major attractions of PS-BPLC devices. In experiment, we compared the decay time of those 12 PSBP samples because the rise time depends on the applied voltage, which is more complicated. The decay time was measured between 90% and 10% transmittance change at 25 °C. The measured decay time of PSBP-1 (without diluter) is 1700 μ s. After adding diluters, the decay time is decreased by 2X–3X, as listed in Table II.

To explain this dramatic change in response time, we refer to Gerber's model [14]:

$$\tau = \frac{\gamma_1' p^2}{k(2\pi)^2}.\tag{8}$$

Here γ'_1 represents the rotational viscosity of PSBP. In our samples with 6 wt% diluters, the variation of p is unnoticeable. The average elastic constant k of PS-BPLC becomes more complicated, since it involves the strong anchoring energy provided by the polymer network and also the interaction between LCs and the chiral dopant. When polymer network is involved, the elastic constant could be 1000X larger than that of the LC host [41]. The diluter effect on the elastic constant of the PSBP is not as significant as the polymer network effect. On the other hand, the viscosity tends to increase noticeably after adding the bulky chiral dopant to the LC host for a PS-BPLC. The small diluter molecules could break the rigid arrangement of the BPLC mixture, which in turn reduces the viscosity effectively and hence the response time.

To eliminate the effect of k, the FoM (defined as $K/\tau \propto \Delta n \cdot \Delta \varepsilon / \gamma'_1$) [29] of the 12 PSBP samples are calculated for comparing the performance of PS-BPLC materials. To see the difference more clearly, we normalized the FoM to that of PSBP-1. From Table II, the FoM can be improved by 1.3X–2.9X with different diluters. Specifically, our data show that the γ'_1 of PSBP-2 is 3.4X lower than that of PSBP-1 (without any diluter). Plugging these values to (8), we find k is ~15% smaller after adding 5CC3, which proves that the dramatic decrease of response time results from the significant decrease in viscosity.

To investigate the activation energy, which is related to the viscosity of the PS-BPLC, we measured the temperature dependent decay time of PSBP-1 and PSBP-2. Results are shown in



Fig. 4. Temperature dependent decay time of PSBP-1 and PSBP-2. Dots are measured data and lines are fitting with (8).

TABLE III INFLUENCE OF DILUTER CONCENTRATION ON THE PS-BPLC PROPERTIES

5CC3 ratio	<i>K</i> (nm/V ²)	Decay time (τ, μs)	FoM (Κ/τ)	<i>T_{m_BP}</i> (°C)	<i>Т_{с_ВР}</i> (°С)
0	8.27	1700	1	5.6	82.3
6%	8.04	573	2.90	7.9	81
10%	7.50	516	3.00	10.4	80.7
15%	7.10	383	3.83	22	79.8

Fig. 4. The decay time decreases as the temperature increases, as described by following equation [42]:

$$\tau \approx B \cdot \frac{\exp(E_a/k_B T)}{(1 - T/T_{c_BP})^{\beta}}$$
(9)

where *B* is a proportionality constant, E_a is the activation energy of the PSBP, and T_{c_BP} is the clearing point of the PS-BPLC. The material constant β has been obtained from the LC host (listed in Table I). From fittings, we obtained $E_a = 738$ meV and $B = 4.0 \cdot 10^{-10} \ \mu s$ for PSBP-1, and $E_a = 573$ meV, $B = 7.8 \cdot 10^{-8} \ \mu s$ for PSBP-2. As expected, PSBP-2 has lower activation energy than PSBP-1, since the diluter in PSBP-2 helps to make the PS-BPLC less viscous. Moreover, PS-BPLC has a higher activation energy than the nematic LC host, indicating that the added chiral dopant indeed increases the viscosity.

V. CONCENTRATION EFFECT

Higher concentration of diluter helps to further reduce the viscosity, but it could have some side effects, such as poor solubility when the diluter has smectic phase, smaller Kerr constant and reduced T_{c_BP} . We prepared another two PSBP samples with 10 wt% and 15 wt% of 5CC3. The decay time decreases significantly with more diluters, as listed in Table III, however the Kerr constant also decreases noticeably. Moreover, T_{m_BP} increases a lot with increased 5CC3 concentration, which is unfavorable for practical applications. Therefore, for a given diluter there exists an optimal concentration. Mixing diluters at a proper ratio is also a common practice for nematic LCs. We could do the same for BPLC in order to increase its solubility.

VI. CONCLUSION

We have investigated the effects of different diluters on PS-BPLC from various aspects. A small amount of diluter will slightly decrease the birefringence and dielectric anisotropy but dramatically reduce the viscosity of the LC host. By doping 6% of diluter, the decay time of the PS-BPLC could be reduced by $2\times-3\times$ while keeping the Kerr constant unchanged. We also studied the diluter effects on the relaxation frequency, melting point and clearing point. By increasing the concentration of a small molecular weight diluter (or a mixture among diluters), the response time of the BPLC can be further decreased, but some undesirable side effects (e.g., smaller Kerr constant) would became noticeable. Therefore, an optimal concentration of diluter should be considered for optimizing the overall performance of the BPLC composite.

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