Supplementary Information

Thermally Reversible Full Color Selective Reflection in a Self-organized Helical Superstructure Enabled by a Bent-core Oligomesogen Exhibiting Twist-bend Nematic Phase

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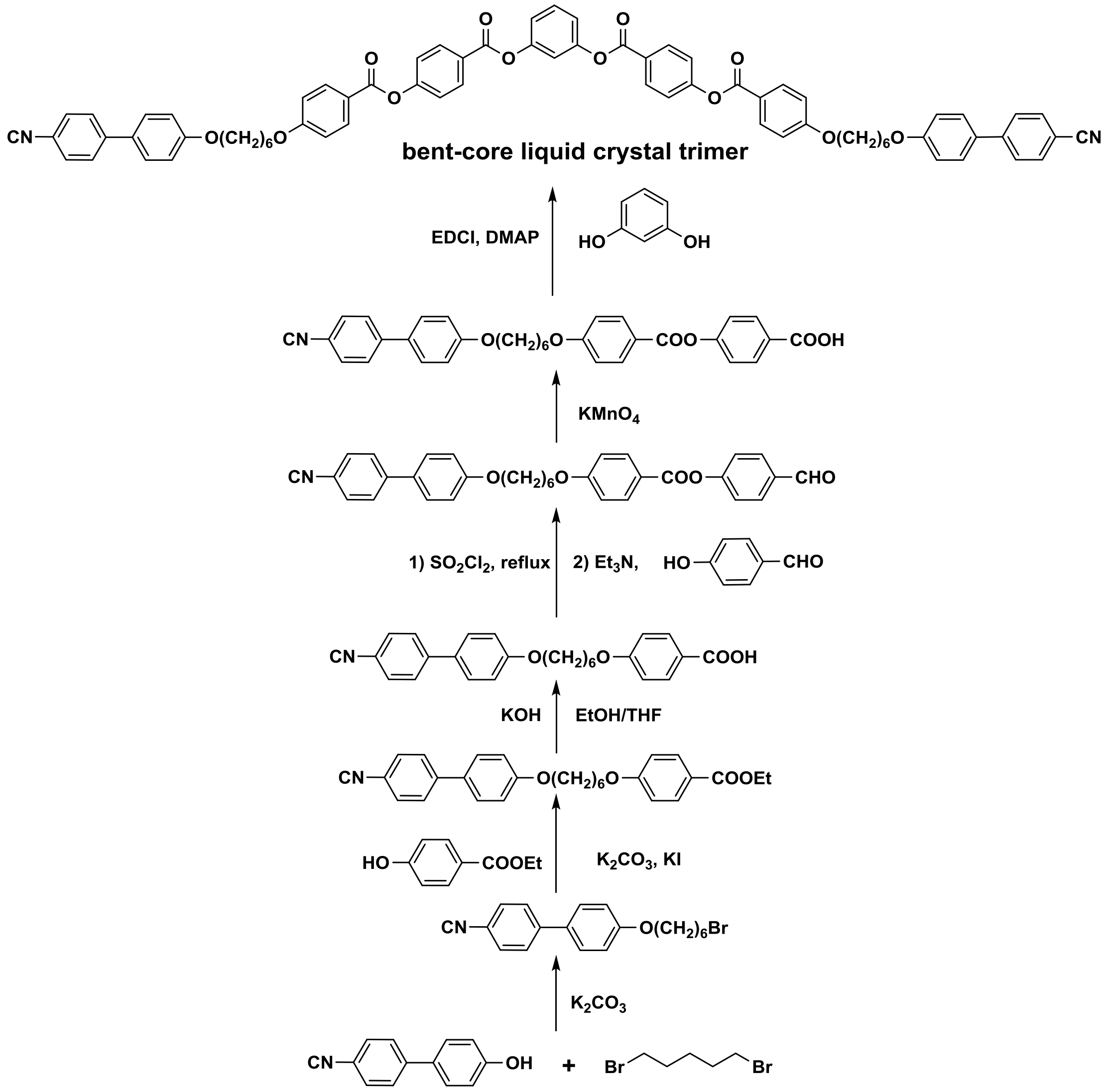
1. Materials and Methods

All starting material, solvents and reagents were purchased from Sigma-Aldrich company and used without further purification. Colunm chromatography was carried out on silica gel (230-400 mesh). Analytical thin layer chromatography (TLC) was performed on commercially coated 60 mesh F254 glass plate. ^{1}H NMR and ^{13}C NMR spectra were recorded on a Bruker 400 spectrometer. Chemical shifts are reported in units (ppm) with the residual sovlent ($CDCl_3$) peak as internal standard. Elemental analysis was performed by Roberston Microscopy Inc. Transition temperatures were taken at the maximum of transition peaks. The optical textures were captured by high-resolution CCD-equipped optical polarizing microscope (Leitz LABORLUX 12 POL, Germany). The reflective spectrum is detected by fiber-connected spectroscope (Ocean Opics USB 2000+). Freeze-fracture transmission electron microscopy (FF-TEM) experiment: The sample was heated to isotropic phase and cooled to 50°C slowly to getthe N_{tb} phase.The sample was then plunge frozen in liquid nitrogen and fractured at $T = -150\,^{\circ}\mathrm{C}$ in a BalTec BAF060 freeze fracture apparatus. The fractured surface was replicated by the deposition of Pt/C (4 nm in thickness) at 45°, followed by the deposition of a 20 nm C film. The replica was collected on carbon coated TEM grids and observed using a FEI Tecnai F20 TEM.

2. Synthesis of Bent-core Liquid Crystal Trimer 1.



Scheme S1. Synthetic route of twist-bend trimer **1**

3. Freeze-fracture transmission electron microscopy (FF-TEM) of the timer 1

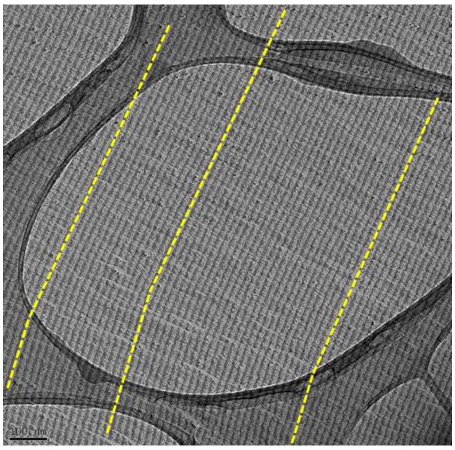


Figure S1. FF-TEM image of N_{tb} trimer **1**. The yellow dashed lines track the curved layers.

4. Reflection band-shifting of $M_{0\sim6}$ with the rising of temperature

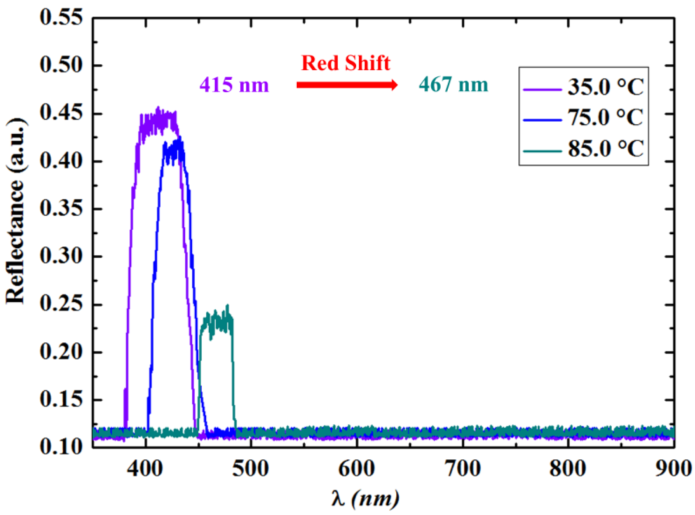


Figure S2. Reflection band shift of the pure CLC $\mathbf{M_0}$ (without trimer **1**).

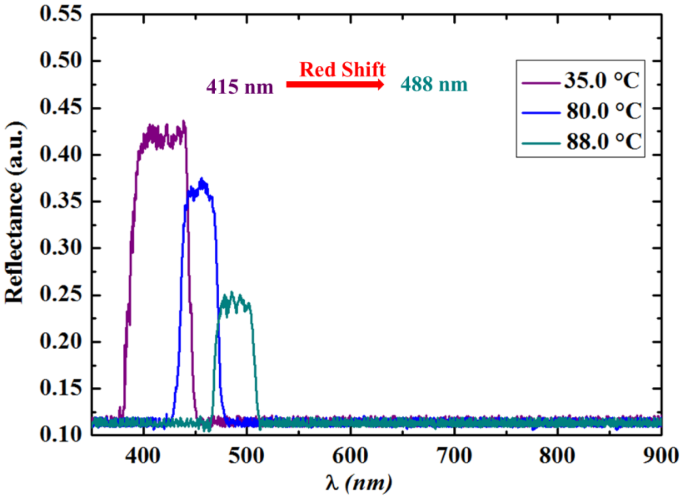


Figure S3. Reflection band shift of the CLC mixture $\mathbf{M_1}$ with 2 wt% of trimer **1**.

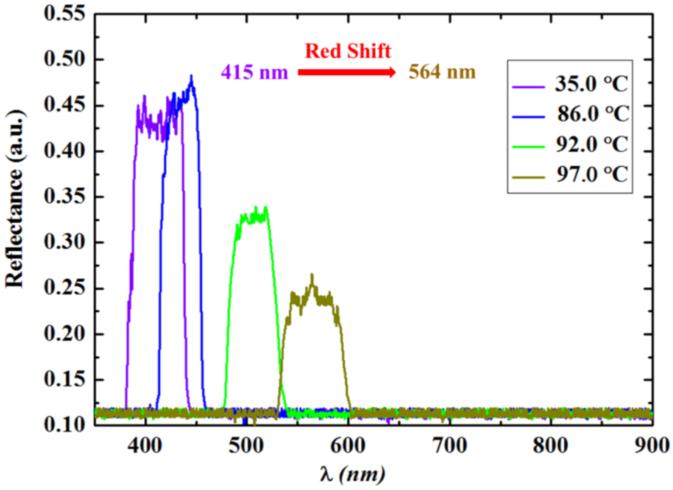


Figure S4. Reflection band shift of the CLC mixture $\mathbf{M_2}$ with 6 wt% of trimer **1**.

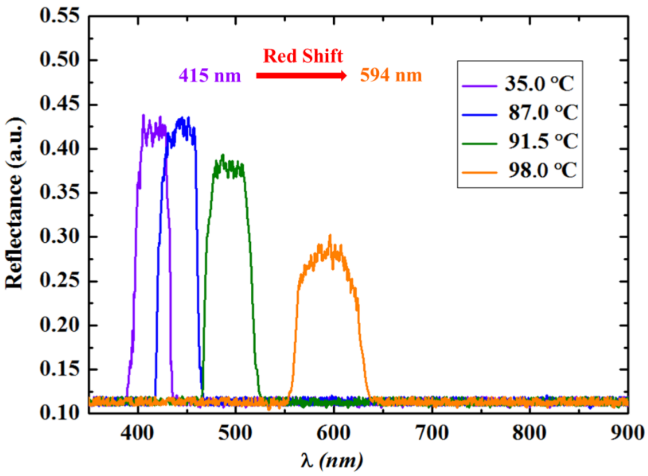


Figure S5. Reflection band shift of the CLC mixture $\mathbf{M_3}$ with 8 wt% of trimer **1**.

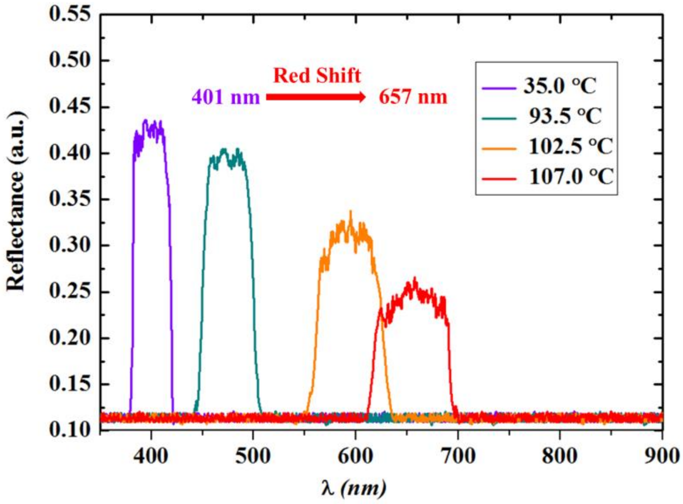


Figure S6. Reflection band shift of the CLC mixture $\mathbf{M_4}$ with 10 wt% of trimer **1**.

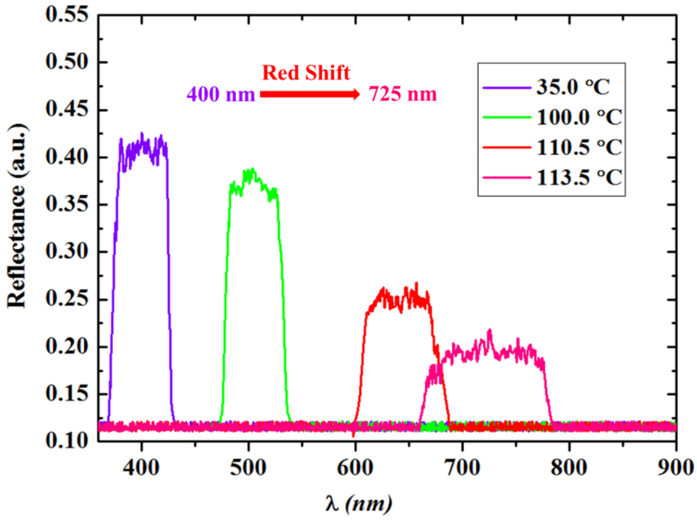


Figure S7. Reflection band shift of the CLC mixture $\mathbf{M_5}$ with 12 wt% of trimer **1**.

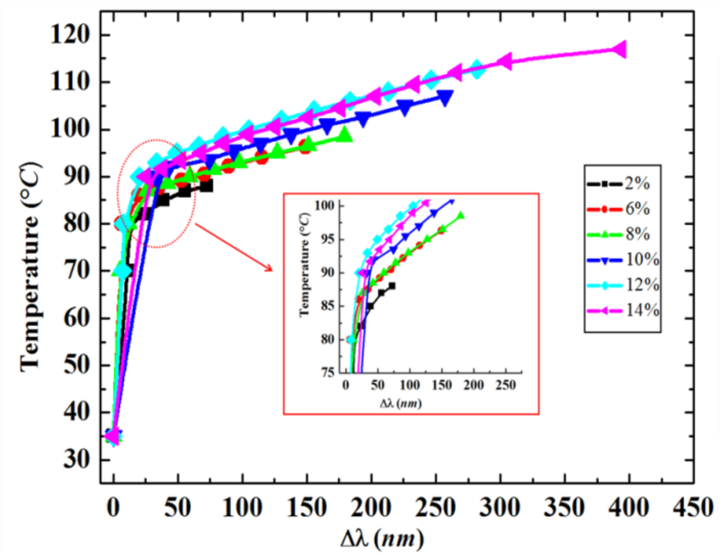


Figure S8. Correlation between temperature and wavelength shifting of mixtures $\mathbf{M_{1\sim6}}$. The starting temperature appearing the obvious band-shift for every sample is almost within the same temperature range (85~90 ^{o}C) as denoted by red-dashed ring.

5. Refractive index and birefringence of sample $\mathbf{M_b}$ (nematic LC and trimer 1)

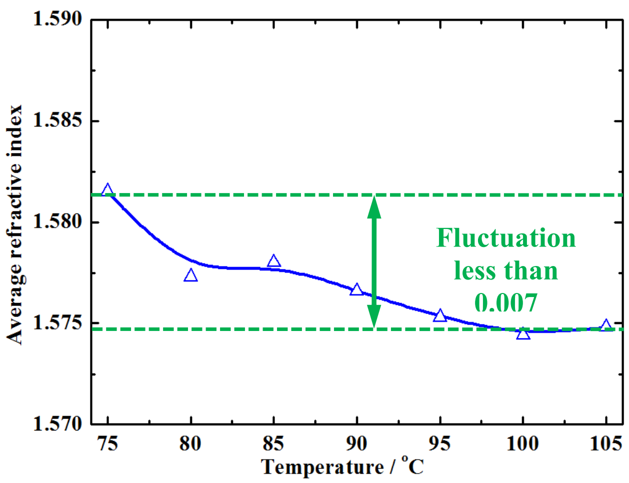


Figure S9. Correlation between the average refractive index of $\mathbf{M_b}$(nematic LC and 14 wt% trimer)and temperature.

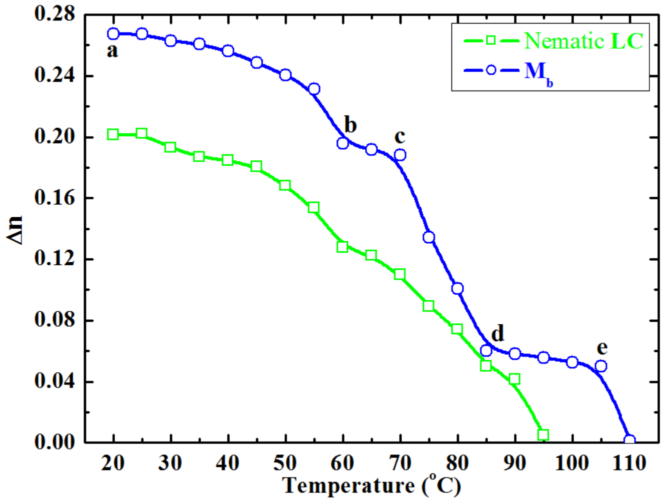


Figure S10. Relationship between birefringence Δn and ambient temperature in M_b (nematic LC and 14 wt% trimer) and pure nematic LC, respectively.

6. Analysis for the relationship of P_0/P-C_{tri}

As shown in Figure 3a and b, the slope of the line is defined as a constant—helical twisted power (*HTP*) of the corresponding chiral dopant, *i.e.*, $1/P = HTP \cdot C$. Herein, the concentration of chiral dopant should not be very large.

In the situation of high temperature, provided that the slope at the linear relationship is a constant *k*, thus,

$$\frac{P}{P_0} = k \cdot C_{tri} \tag{1}$$

where P_0 is the original pitch length without trimer dopant. Therefore, P_0 can be expressed by the helical twisted power of chiral dopant (HTP_0), and its concentration (C_{chiral}),

$$\frac{1}{P_0} = HTP_0 \cdot C_{chiral} \tag{2}$$

By combining Eqs. (1) and (2), the pitch length of a system with trimer dopant is

$$\frac{1}{P} = \frac{HTP_0}{k} \cdot \frac{C_{chiral}}{C_{tri}} = [HTP] \cdot [C] \tag{3}$$

$$[HTP] = \frac{HTP_0}{k}; \quad [C] = \frac{C_{chiral}}{C_{tri}} \tag{4}$$

in which, [*HTP*] and [*C*] are defined respectively as the modulated helical twisted power and the modulated concentration of chiral dopant of the trimer doped system. The corresponding modulation parameters are *k* and C_{tri}. Here, *k* is determined by molecular interaction between

LCs and trimer molecules, and it is related to the molecular interaction. As shown in Figure 3b, for a fixed temperature k is a constant.

Equation (3) indicates that the helical pitch at a certain temperature of trimer-doped system can be expressed in the same form that a conventional chiral-doped system. The trimer **1** induces a weakened effect of helical twisted power, however such effect is only dependent on the aforementioned molecular interaction, *i.e.*, HTP-modulation parameter—*k*, thus the modulated [*HTP*] is a constant at a certain temperature. From Figure 3b, as the temperature is increased, k gets larger and therefore the pitch length increases, thus explaining the observed red shift.

On the other hand, in the situation of low temperature, a non-linear relationship of P/P_0 and C_{tri} was shown in Fig. 3a, and can be expressed as,

$$\frac{P}{P_0} = \frac{1}{a + b \cdot e^{c \cdot C_{tri}}} \tag{5}$$

where a, b and c are fitting parameters.

Substituting Eq. (2) into Eq. (5), we obtain the expression of $1/P$,

$$\frac{1}{P} = a \cdot HTP_0 \cdot C_{chiral} + b \cdot HTP_0 \cdot e^{c \cdot C_{tri}} \cdot C_{chiral} \tag{6}$$

where $a+b=1$ (because $P_0=P$ when $C_{tri}=1$) and "$e^{c \cdot C\mathrm{tri}}$ " is a non-linear factor which influences the HTP of the system. In Eq. (6), "c" is an interaction parameter determined by the molecular structure, solubility and the temperature. According to Eq. (6), when the temperature is invariable, with the increasing of trimer **1**, the HTP of the system is strengthened. This HTP strengthening has been demonstrated in our experiment and the previous publications. In addition, as we mentioned in the manuscript, the bent-core having similar bent-angle and molecular length with the bent-core fragment of trimer **1** was doped into the same CLC host to repeat the same experiment, the similar chirality enhancement was observed, confirming the effect of bent-core fragment on the chirality enhancement, a fact that has been observed in other systems elsewhere.