

Supporting Information for

**Thermally Reconfigurable Janus Droplets with Nematic
Liquid Crystalline and Isotropic Perfluorocarbon Oil
Compartments**

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Thermodynamics of phase coexistence

Our observations of coexisting nematic and isotropic phases (as shown in Figure 3 of the main manuscript) is consistent with the Gibb's phase rule. The number of independent compositional degrees of freedom (C) is 2 for perfluorobenzene (FB) and E7 (E7 is a mixture of 4 components, but they are not independent because the composition of E7 is constant). The number of phases (P) is 2 (nematic and isotropic). It follows that the degrees of freedom (F) are equal to $C+2-P=2$, thus allowing the existence of coexisting nematic and isotropic phases.

For nematic (N) and isotropic (I) phases at equilibrium, the solute (FB) chemical potential in each of the two phases must be equal¹:

$$\gamma^N C_{FB}^N = \gamma^I C_{FB}^I$$

where γ^N and γ^I are activity coefficients of the solute in the nematic phase and isotropic phase, respectively, and C_{FB}^N and C_{FB}^I are the concentrations of solute. Accordingly,

$$\frac{C_{FB}^I}{C_{FB}^N} = \frac{\gamma^N}{\gamma^I},$$

Inspection of Figure 3 of the main manuscript reveals that $\frac{\gamma^N}{\gamma^I}$ is larger for FB than toluene in the two-phase system based on E7. We speculate that this difference arises from electrostatic interactions between FB and the aromatic components of E7 that are absent for toluene (see main text for details).

1 G. R. Luckhurst and G. W. Gray, eds., *The Molecular Physics of Liquid Crystal*, Academic Press, 1979.

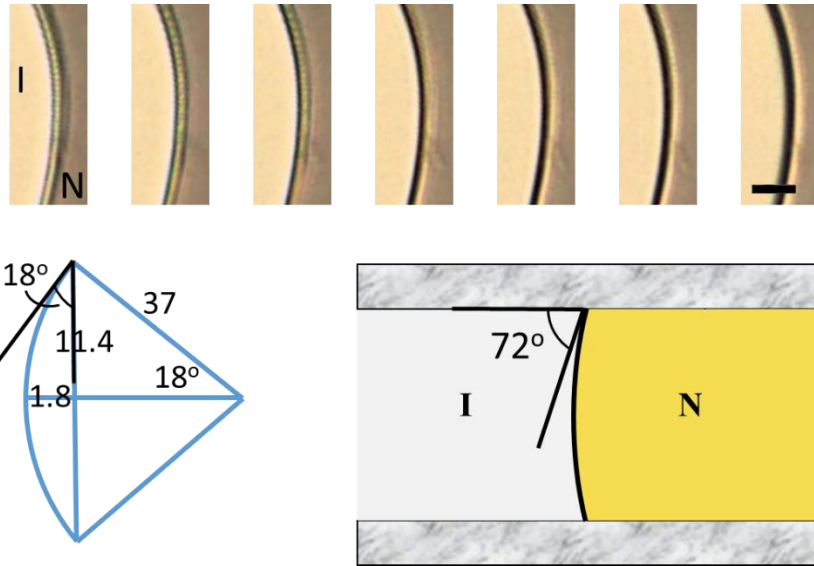


Fig. S1 Curvature of the N-I interface. Top row are micrographs of a N-I interface that expands both glass surfaces of an optical cell with changing focal plane from top to bottom of a FB-E7 ($C_{FB}=5\%$ v/v) mixture at $44.0\text{ }^{\circ}\text{C}$ (Nematic-Isotropic coexistence). Scale bar, $10\text{ }\mu\text{m}$.

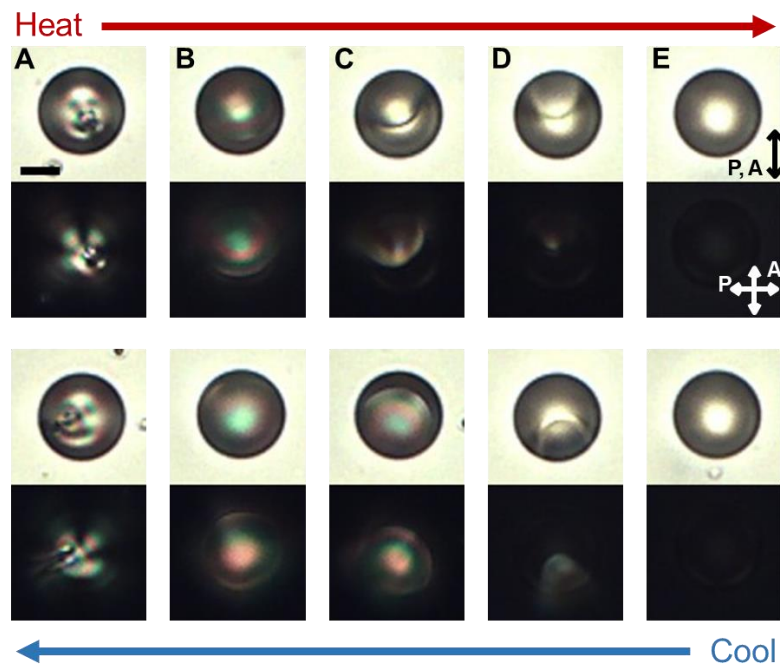


Fig. S2 FB-E7 ($C_{\text{FB}}=5\%$ v/v) droplet in $100\ \mu\text{M}$ SDS aqueous solution. (A-E) Top and bottom rows are micrographs of a LC droplet upon heating and cooling across the N-I biphasic region at (A) $30.0\ \text{°C}$ (Nematic), (B) $42.5\ \text{°C}$, (C) $43.5\ \text{°C}$, (D) $44.3\ \text{°C}$, (E) $45.0\ \text{°C}$ (Isotropic), respectively. Scale bar, $10\ \mu\text{m}$.

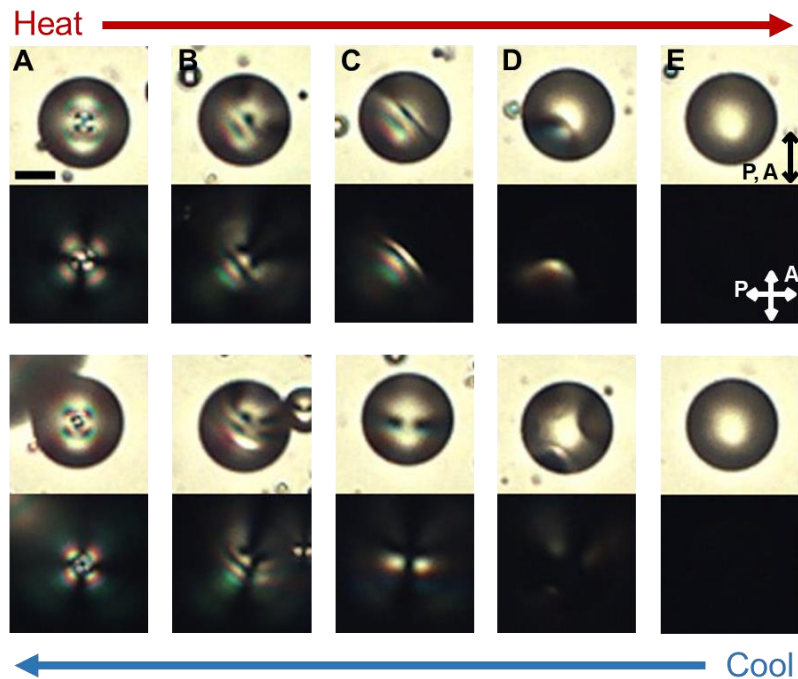


Fig. S3 FB-E7 ($C_{FB}=5\%$ v/v) droplet in 1 mM SDS aqueous solution. (A-E) Top and bottom rows are micrographs of a LC droplet upon heating and cooling across the N-I coexisting region at (A) 35.0 °C (Nematic), (B) 45.0 °C, (C) 46.0 °C, (D) 47.0 °C, (E) 47.5 °C (Isotropic), respectively. Scale bar, 10 μm .

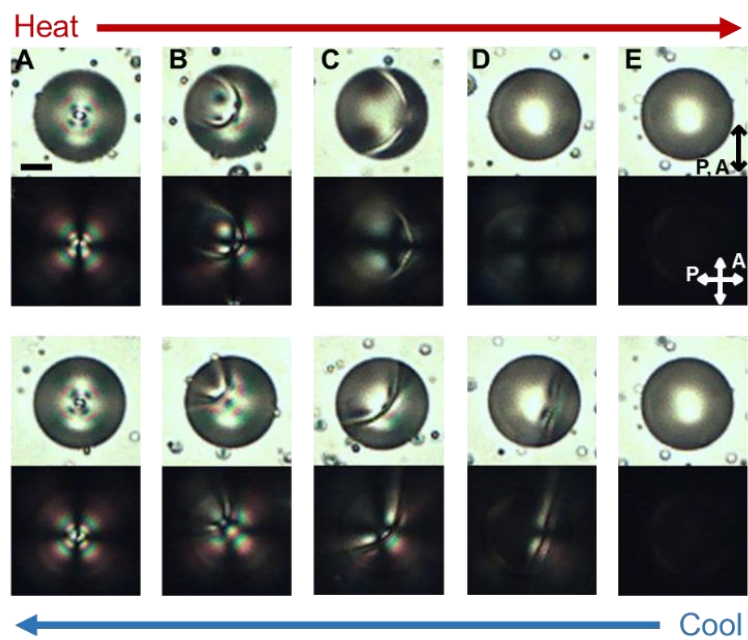


Fig. S4 FB-E7 ($C_{FB}=5\%$ v/v) droplet in 2 mM SDS aqueous solution. (A-E) Top and bottom rows are micrographs of a LC droplet upon heating and cooling across the N-I biphasic region at (A) 41.0 °C (Nematic), (B) 42.3 °C, (C) 43.0 °C, (D) 43.8 °C, (E) 45.0 °C (Isotropic), respectively. Scale bar, 10 μm .

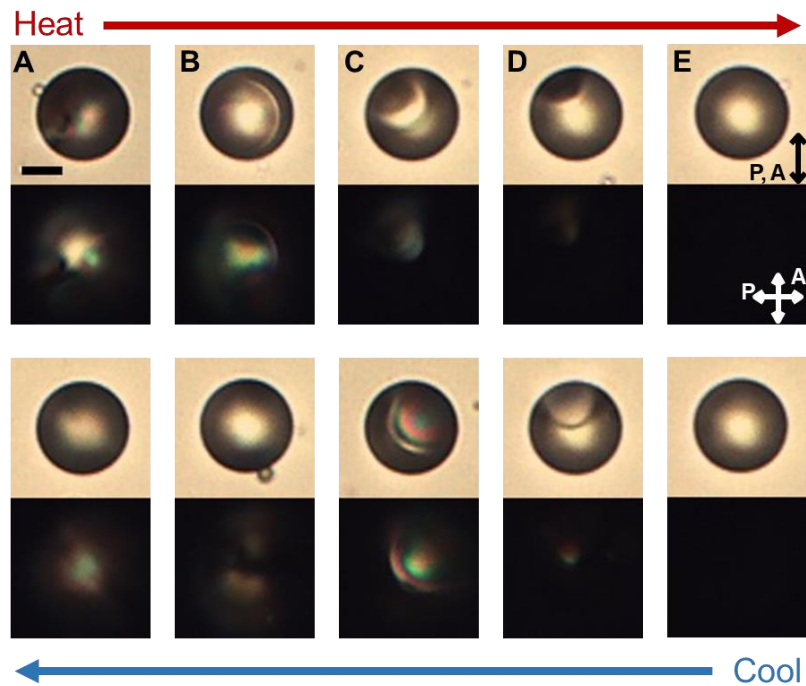


Fig. S5 FB-E7 ($C_{FB}=5\%$ v/v) droplet in 0.5 mM PFOA aqueous solution. (A-E) Top and bottom rows are micrographs of a LC droplet upon heating and cooling across the N-I biphasic region at (A) 35.0 °C (Nematic), (B) 40.5 °C, (C) 41.5 °C, (D) 42.0 °C, (E) 42.5 °C (Isotropic), respectively. Scale bar, 10 μm .

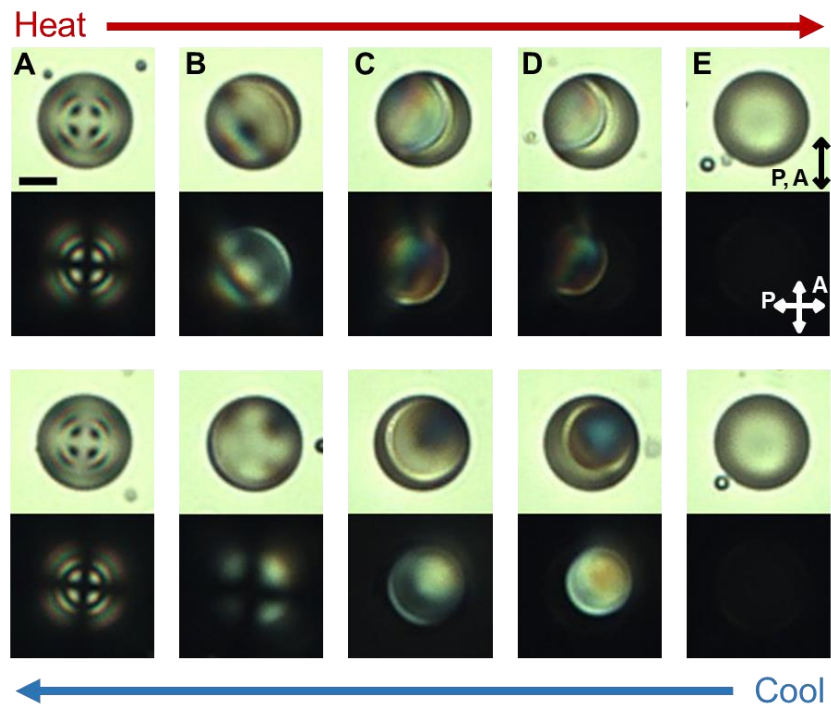


Fig. S6 FB-E7 ($C_{\text{FB}}=5\%$ v/v) droplet in 1 mM PFOA aqueous solution. (A-E) Top and bottom rows are micrographs of a LC droplet upon heating and cooling across the N-I biphasic region at (A) 35.0 °C (Nematic), (B) 44.3 °C, (C) 45.0 °C, (D) 45.5 °C, (E) 46.8 °C (Isotropic), respectively. Scale bar, 10 μm .

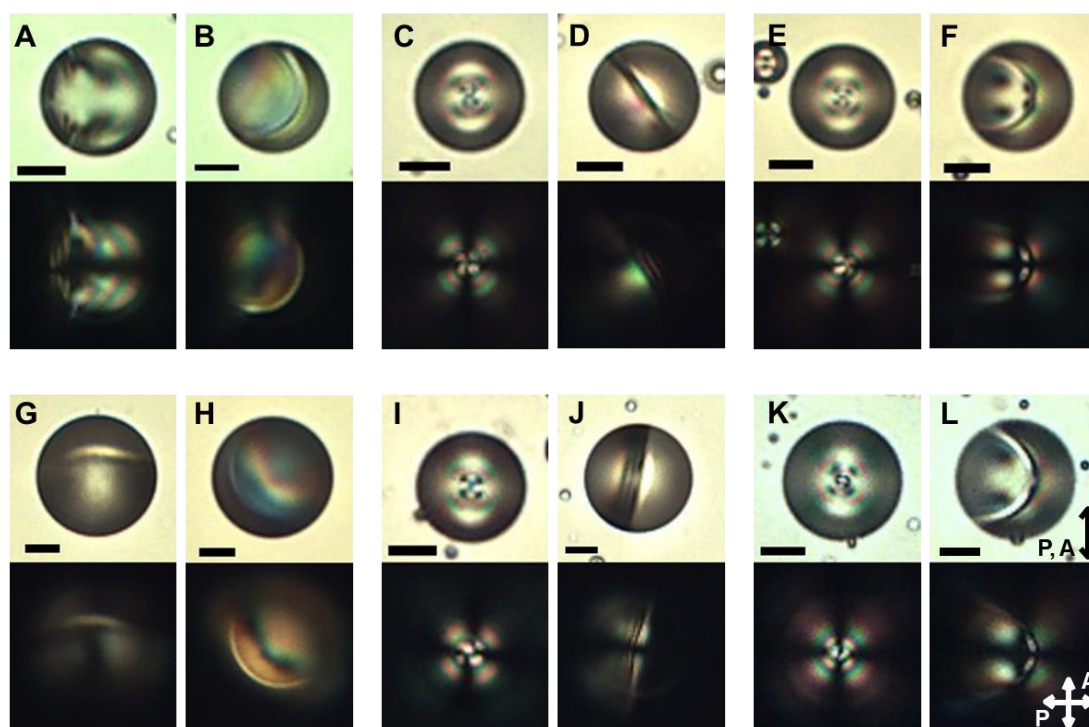


Fig. S7 A comparison of FB-E7 droplets at (A-F) pH = 3 and (G-L) pH = 7. Droplets both (A,C,E,G,I,K) in single phase state and (B,D,F,H,J,K) in biphasic state in the presence of (A,B,G,H) 1 mM PFOA, (C,D,I,J) 1 mM SDS and (E,F,K,L) 2 mM SDS. Scale bar, 10 μ m. pH value was adjusted using sodium hydroxide and hydrochloric acid.

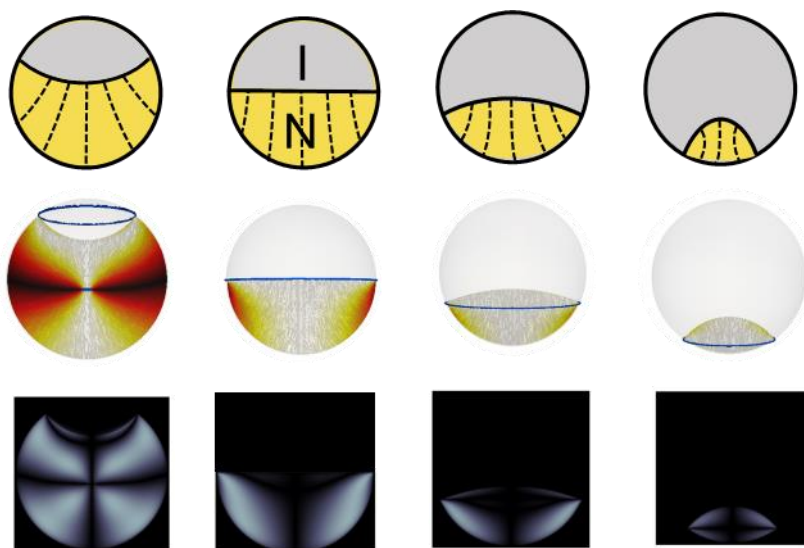


Fig. S8 Simulated schematic illustrations (row 2) and polarized light micrographs (row 3) for multi-compartment droplets under the influence of strong anchoring strength at N-I interfaces. Defect lines (blue loops schematic illustrations) were generated.