

## Supplementary Information

# Colloid-in-Liquid Crystal Gels Formed via Spinodal Decomposition

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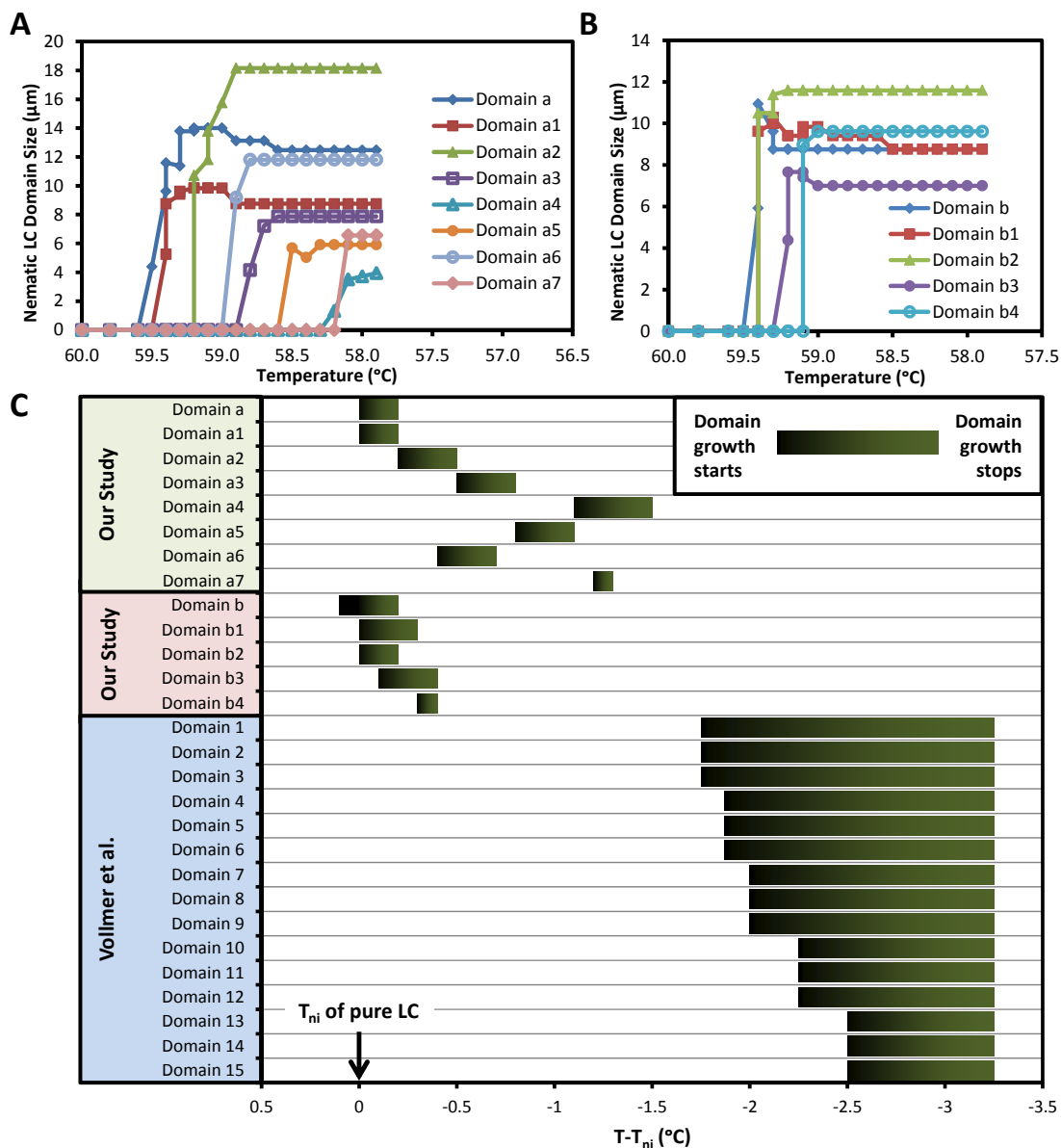
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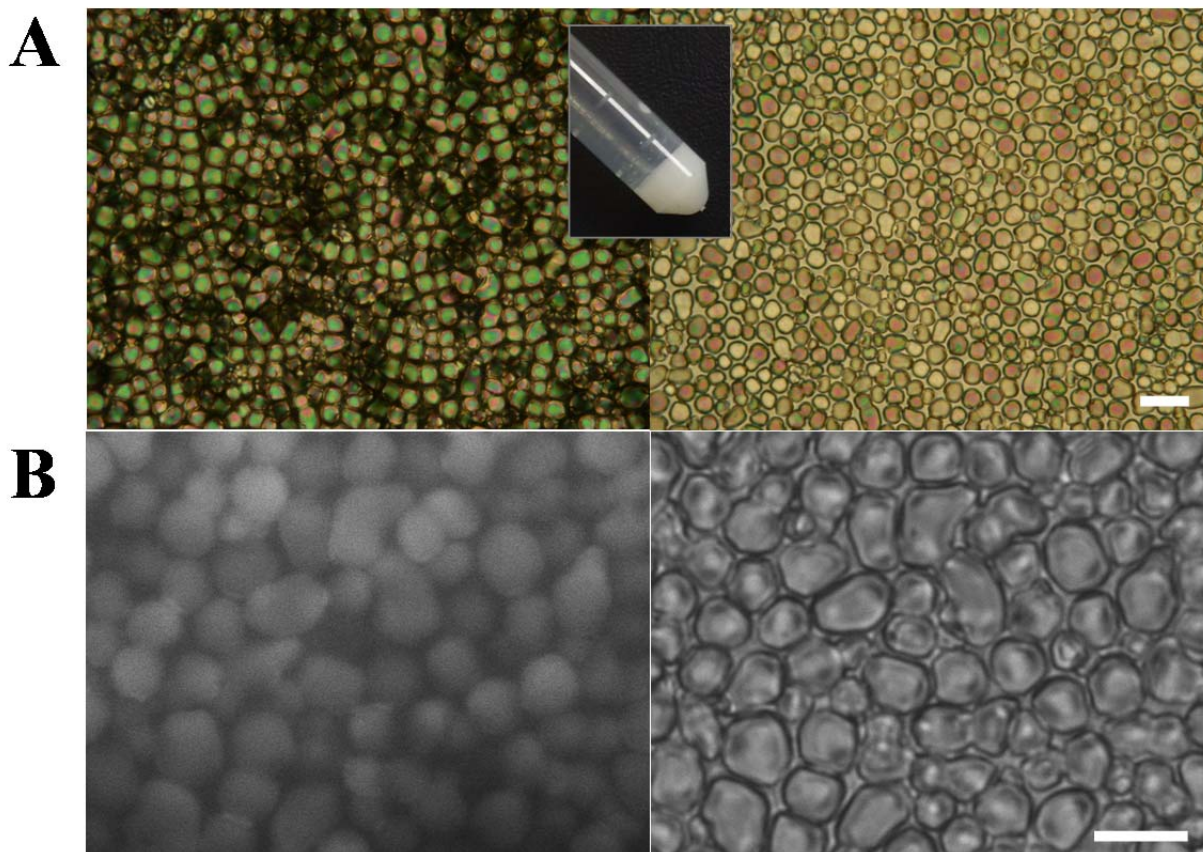
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### **Dynamics of growth of nematic domains during formation of CLC gels**

Figure SI.1 A and B show the growth of nematic domains in PS-colloid/E7 samples as a function of temperature (SI.1A and 1B). Figure SI.1C compares the growth characteristics of the nematic domains in the PS-colloid/E7 samples to the growth characteristics of the nematic domains in the CLC gels described by Vollmer et al.<sup>1</sup>



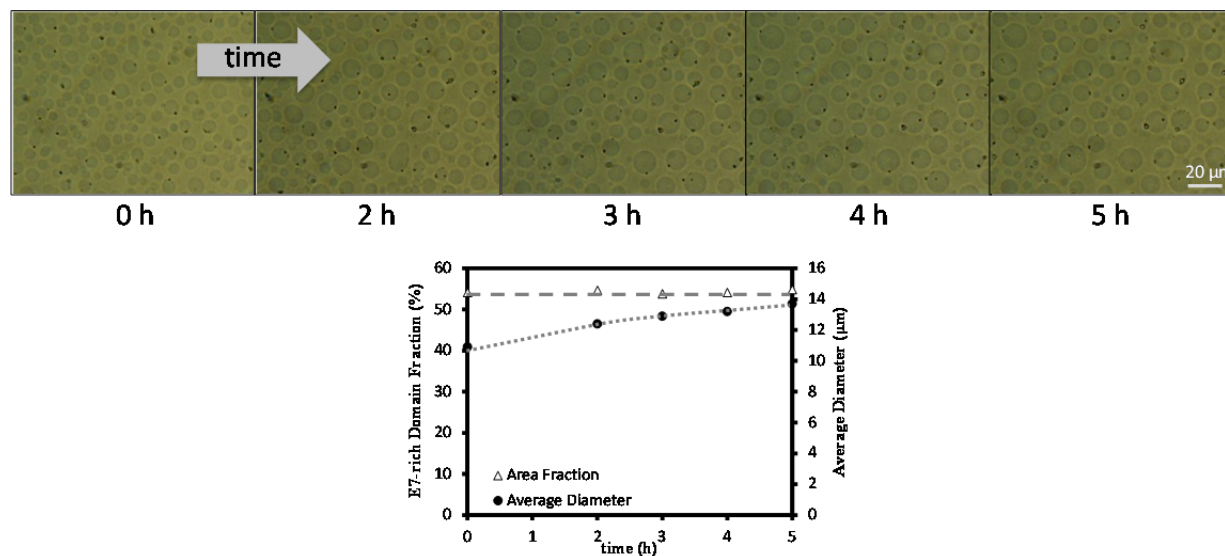
**Figure SI.1.** Growth characteristics of nematic domains during formation of CLC gels. **(A)** PS-colloid/E7 mixtures. Size of Domain *a* of Figure 1 of main text and neighboring domains which are labeled **a1** to **a7**, **(B)** Size of Domain *b* of Figure 1 of main text and neighboring domains labeled as **b1** to **b4** **(C)** Comparison of growth characteristics of nematic domains observed with PS-colloid/E7 mixtures to the growth characteristics of nematic domains of PMMS/5CB mixtures reported in Figure 2 of Vollmer et al.<sup>1</sup>



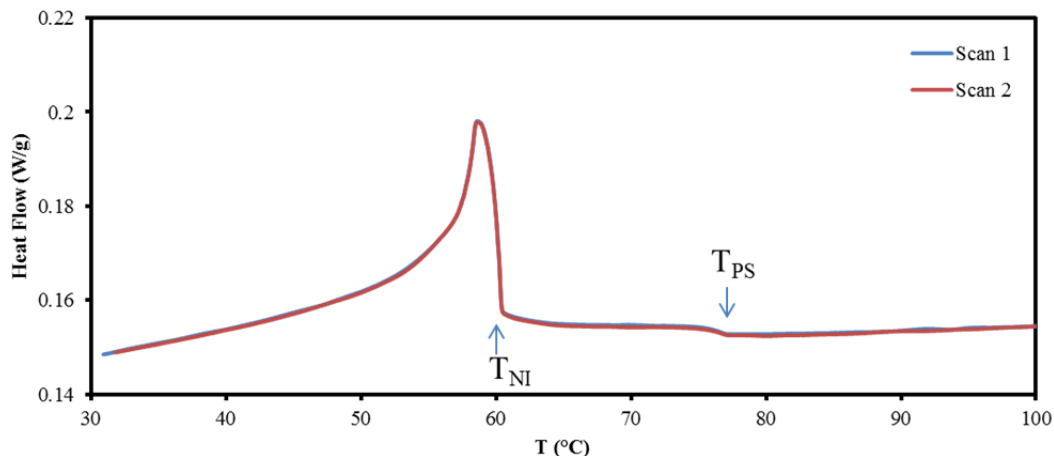
**Figure SI.2.** (A) Polarized light (left) and corresponding brightfield (right) images of a 13.3 wt% PS-colloid/E7 mixture at room temperature. (B) Fluorescence (left) and corresponding brightfield images (right) of a 13.3 wt% PS-colloid/E7 mixture doped with 0.1% Nile Red at room temperature. (Scale bars correspond to 30  $\mu\text{m}$ )

## Dynamics of Evolution of Area Fraction of Phases and Domain Size

A thin film of 13.3 wt% PS-colloid/E7 gel was heated to 80.0°C, and then cooled at a rate of 0.2°C/min to 63.0°C (same cooling rate as used in Figure 1) at which temperature the system formed a cellular microstructure. Subsequently, images were recorded while holding the sample at a constant temperature of 63.0°C for ~5 hours (**Figure SI.3**).



**Figure SI.3.** Isothermal time-lapse brightfield images of a mixture of 13.3 wt% PS-colloids and E7. The images were recorded after the sample had been cooled at a rate of 0.2°C/min to 63°C (at t=0 h). The plot in the lower panel shows the area fraction of the E7-rich phase and the average size of E7-rich domains.

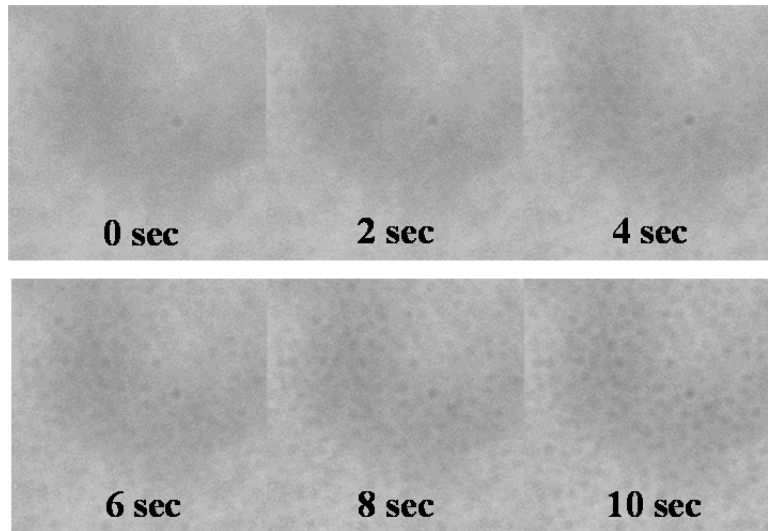


**Figure SI.4.** Duplicate DSC scans of a 13.3 wt % PS-colloid/E7 sample. The sample was cooled at 5.0 °C/min. The temperature at which phase separation of the colloids in isotropic E7 was observed is indicated as  $T_{PS}$ .

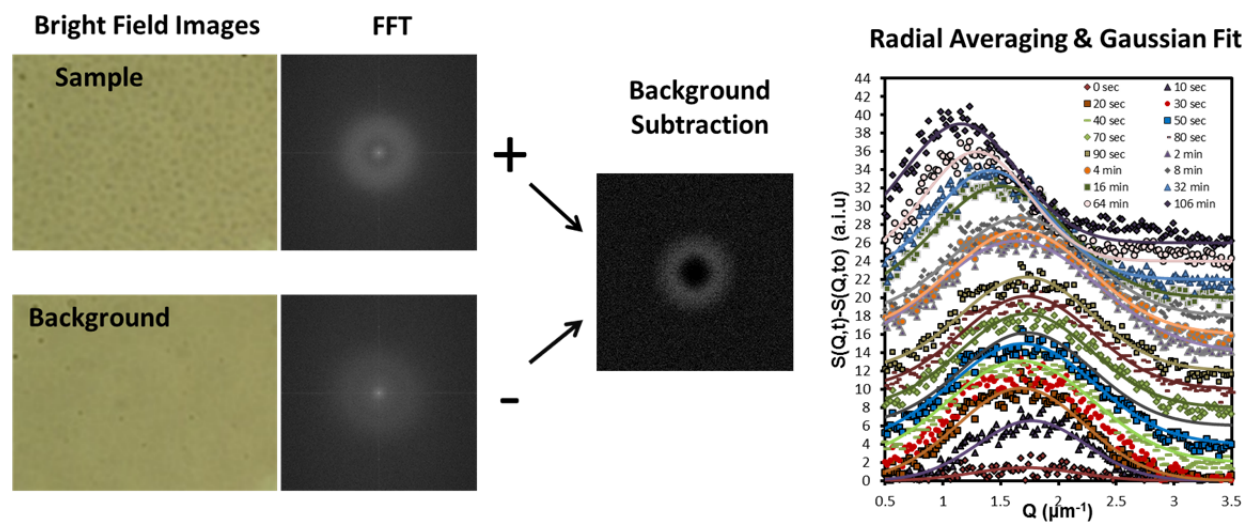
From the DSC scan shown in Figure SI.4, we determined  $T_{NI}$  of a 13.3 wt% PS-colloid/E7 sample to be  $\sim 59.5^{\circ}\text{C}$ . This value is in good agreement with the value of  $T_{NI}$  determined by optical microscopy ( $59.4^{\circ}\text{C}$ ). In addition to  $T_{NI}$ , we observed a small exothermic process to occur at a temperature that coincided with the onset of the phase separation of the PS-colloids in the isotropic phase of E7. We note that the temperature at which the onset of phase separation of the PS-colloids was observed in E7 was dependent on the thickness of the sample. When using thick samples (thickness greater than  $20\mu\text{m}$ ), phase separation of the PS-colloids in E7 was determined by microscopy to occur at  $76^{\circ}\text{C}$  (for 13.3 wt% sample). This temperature coincides with the exothermic signature in the DSC thermogram shown in Figure SI.4.

### Images of initial stage of phase separation

The brightfield optical micrographs in Figure SI.5 show the initial stages of phase separation of a 13.3 wt% PS-colloid/E7 mixture after the system was quenched to 67.0°C.



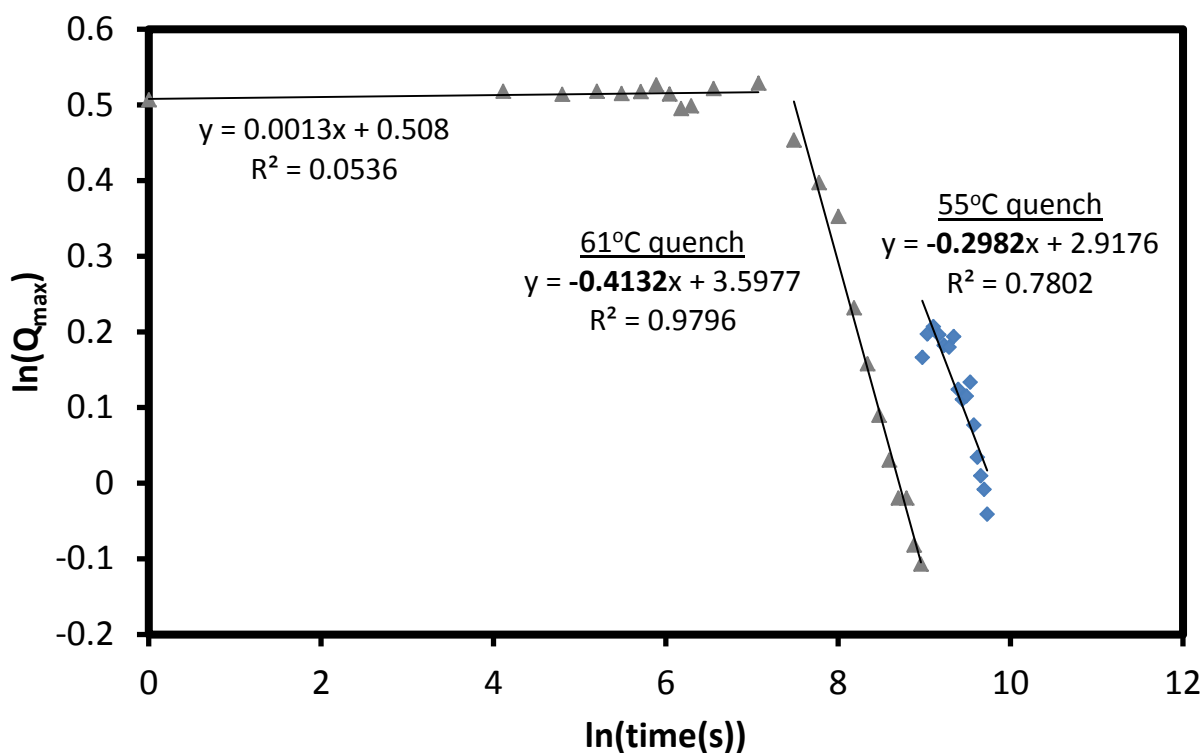
**Figure SI.5.** Time-lapse images of a 13.3 wt% sulfate-PS/E7 mixture quenched to 67.0°C from 71.0°C.



**Figure SI.6.** Illustration of the procedure used for the FFT analysis of thin films of 13.3 wt% PS-colloids in E7.

### Characterization of the Coarsening of the Microstructure:

A thin film of a 13.3 wt% sulfate-PS/E7 mixture was quenched sequentially to 61 °C from 80 °C, and then to 55 °C from 61 °C. The images were analyzed as described in Figure SI.6. We note that the depth of the quench used to prepare the sample in Figure SI.7 was greater than that used to prepare the samples that are described in Figure 8. Past studies have demonstrated that deep quenches can result in high rates of coarsening due to “viscous hydrodynamic” mechanisms.<sup>2,3</sup> For this reason, the coarsening seen in Figure 8 cannot be directly compared to Figure SI.7.



**Figure SI.7.** Time-dependence of the structural peak in a 13.3 wt% sulfate-PS/E7 mixture quenched from 80°C to 61 °C and then to 55 °C.



## SI References

1. D. Vollmer, G. Hinze, B. Ullrich, W. C. K. Poon, M. E. Cates and A. B. Schofield, *Langmuir*, 2005, **21**, 4921-4930.
2. E. D. Siggia, *Physical Review A*, 1979, **20**, 595-605.
3. A. J. Bray, *Advances in Physics*, 2008.