Electronic Supplementary Information (ESI)

Positioning Colloids at the Surfaces of Cholesteric Liquid Crystal Droplets

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Simulation details

The numerical parameters used in the simulations reported in this paper were $A = 1.067 \times 10^5$ J/m^3, $U = 5$, $L = 6$ pN, $W = 1 \times 10^{-3}$ J/m^2 for the droplet surface and $W_c = 1 \times 10^{-5}$ J/m^2 for the colloid surface. The droplet diameter D was 2 μm and the colloid diameter was 200 nm, if not otherwise specified. The colloid surface anchoring strength was chosen according to experimental measurements.[1] The ratio of the Kleman-de Gennes length (L/W_c) to the colloid radius (R_p) is 6, which indicates a non-negligible effects of surface-induced order.[2]

Rare cholesteric droplet configurations

Two rarely observed cholesteric droplets configurations are shown in Fig. S1. Fig. S1A shows a droplet with a defect structure along one radius of one hemisphere (there is not defect evident in the other hemisphere). Figs S1B and S1C show experimental and simulation results for a droplet exhibiting a second rare configuration. The droplet appears to exhibit surface defects at the two poles and two bulk defect rings.

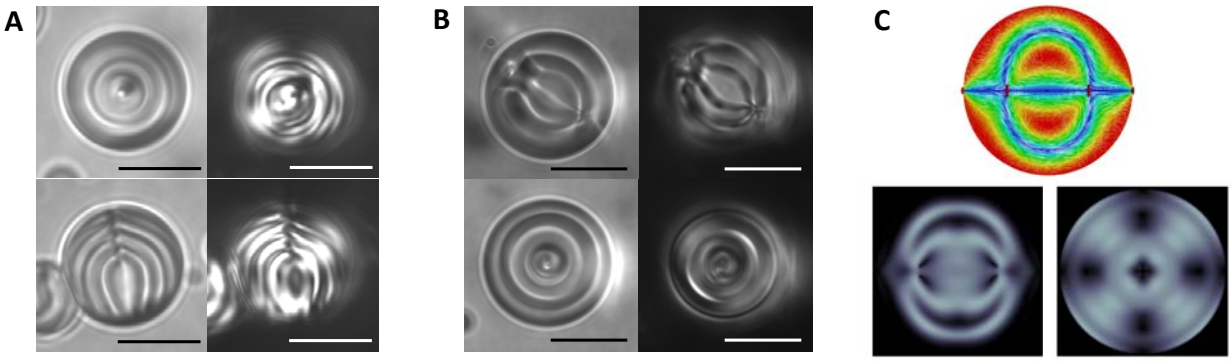

Fig. S1. Bright field and polarized light micrographs of two rarely observed cholesteric droplet structures. Representative micrographs of droplets of (A) 2.5% and (B) 1% wt/wt S-811 in 5CB. (C) Director field of a chiral LC droplet for $N = 3$ that has a similar configuration as in (B). Defects are shown in red (isosurface for $S = 0.5$). Scale bars: (A) 10 μm and (B) 20 μm.

Images of DSS droplet configurations

Here we show images of another representative DSS droplet configuration. We observe alternating dark and bright spots, rather than a single defect line, along the diameter of the droplet. This image is consistent with previous computer-based simulations by Sec *et al*.[3]

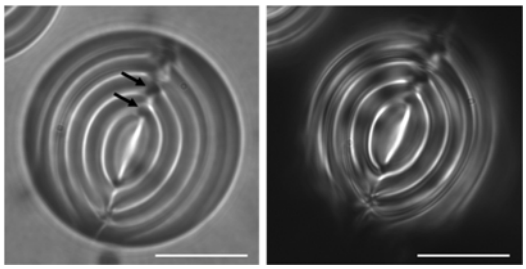

Fig. S2. Bright field and polarized light micrographs of a 1% wt/wt S-811 in 5CB droplet exhibiting a DSS configuration. Arrows indicate two representative LC defects within the droplet. Scale bars: 20 μm.

Influence of *N* on the angle separating two surface defects

For *N* between 3 and 4, we observed the angle separating the two surface defects to decrease with increase in *N*, leading to a continuous change from TBS at *N* = 3 to RSS at *N* = 4, as shown in Fig. S3 and Ref. 4. This transition reflects changes in the bulk elastic and surface anchoring energies that occur with increasing *N*. A more detailed analysis can be found in Ref. 4.

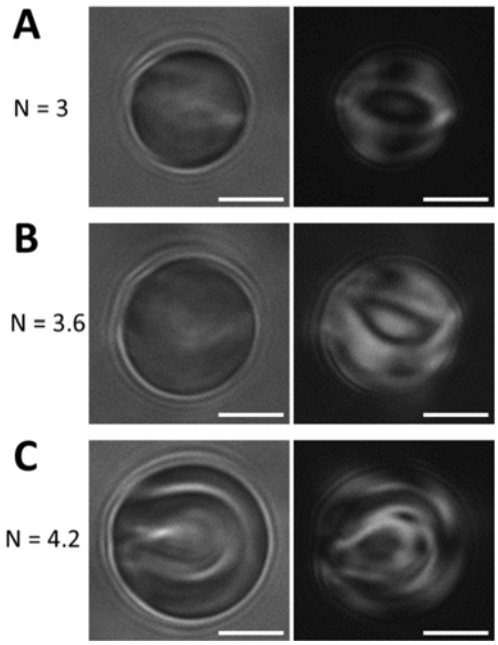

Fig. S3. Size-dependent transition from TBS to RSS for cholesteric droplets containing 1 wt% S-811 in 5CB. Bright field (left) and polarized light (right) micrographs for droplets with (A) *N* = 3, (B) *N* = 3.6, and (C) *N* = 4.2. Scale bars: 5 μm.

Localization of colloids on the surface of TBS and BS droplets

We performed additional experiments to investigate the effects of colloid size on colloid localization on the surfaces of TBS droplets. Fig. S4A shows a plot of the probability of finding 200 nm-in-diameter colloids positioned at an angle ϕ from the surface defects of TBS droplets with diameters ranging from 6 to 10 μm (*N* = 2 to 3.6). Inspection of Fig. S4A reveals that 27% of the

colloids localized at the defects (ϕ = 0°) and 15% were localized at ϕ = 90°. In contrast to results obtained using 1 µm colloids, a significant percentage of the 200 nm-in-diamter colloids localize away from the point defects.

For comparison, we performed experiments using nematic BS droplets. As shown in Fig. S4B, for 1 µm-in-diameter colloids, all of the colloids localize at the point defects, whereas ~ 45% of the 200 nm-in-diameter colloids localize away from the point defects. Overall, these results reveal that the relative positioning behaviors of colloids on TBS and BS droplets is dependent on the size of the colloids: for 1 µm colloids, distinct behaviors were observed, whereas for 200 nm diameter colloids, similar behaviors were exhibited in both chiral and achiral systems.

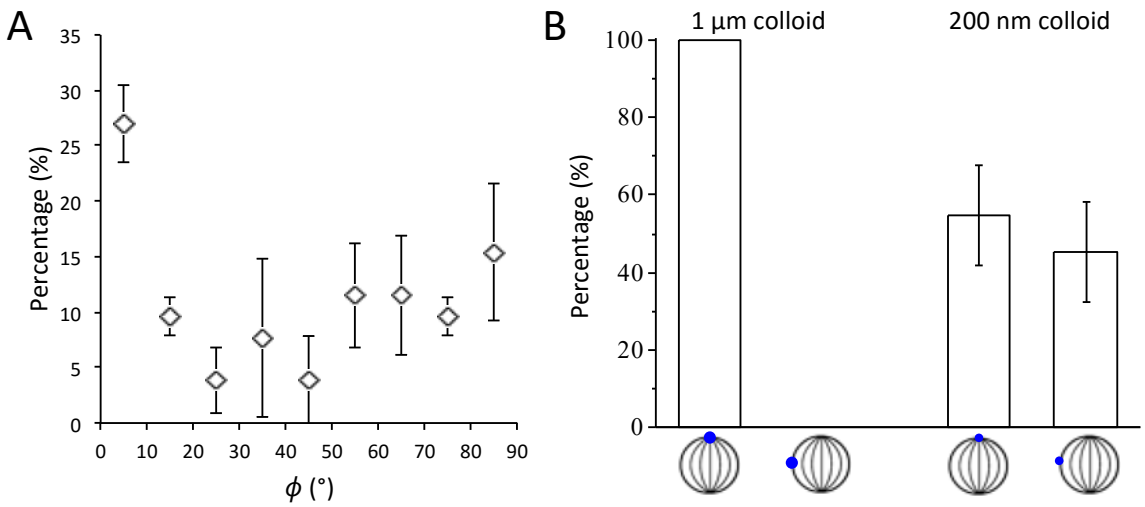

Fig. S4. Statistics of colloid localization on (A) cholesteric TBS and (B) nematic BS droplets. The data in (A) was assembled from a total of 52 droplets with 200 nm-in-diameter colloids and data in (B) was assembled from a total of 33 droplets with 1 µm-in-diameter colloids and 44 droplets with 200 nm-in-diameter colloids (3 independent experiments for each case).

Images of RSSII droplet configurations

Here we show a representative RSSII droplet configuration corresponding to θ = 85°. Video 5 also includes images of the droplets taken while undergoing rotational diffusion.

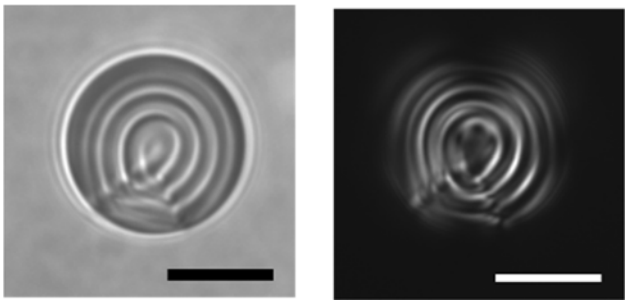

Fig. S5. Bright field and polarized light micrographs of a droplet comprised of 2.5% wt/wt S-811 in 5CB exhibiting a RSSII configuration with θ=85°. Scale bars: 10 μm.

Supporting information video 1: TBS droplet

Supporting information video 2: DSS droplet

Supporting information video 3: RSS droplet

Supporting information video 4: An RSSII droplet heating and cooling experiment.

Supporting information video 5: RSSII droplet

References

1. D. Seo, K. Muroi, T. Isogami, H. Matsuda and S. Kobayashi, *Jpn. J. Appl. Phys.*, 1992, **31**, 2165-2169.
2. M. Kleman and C. Williams, *Philos. Mag.*, 1973, **28**, 725-732.
3. D. Sec, T. Porenta, M. Ravnik and S. Zumer, *Soft Matter*, 2012, **8**, 11982-11988.
4. Y. Zhou, E. Bukusoglu, J. A. Martinez-Gonzalez, M. Rahimi, T. F. Roberts, R. Zhang, X. Wang, N. L. Abbott and J. J. de Pablo, *ACS Nano*, 10.1021/acsnano.6b01088.