

## **Supporting Information**

# **Spontaneous Formation of Water Droplets at Oil-Solid Interfaces**

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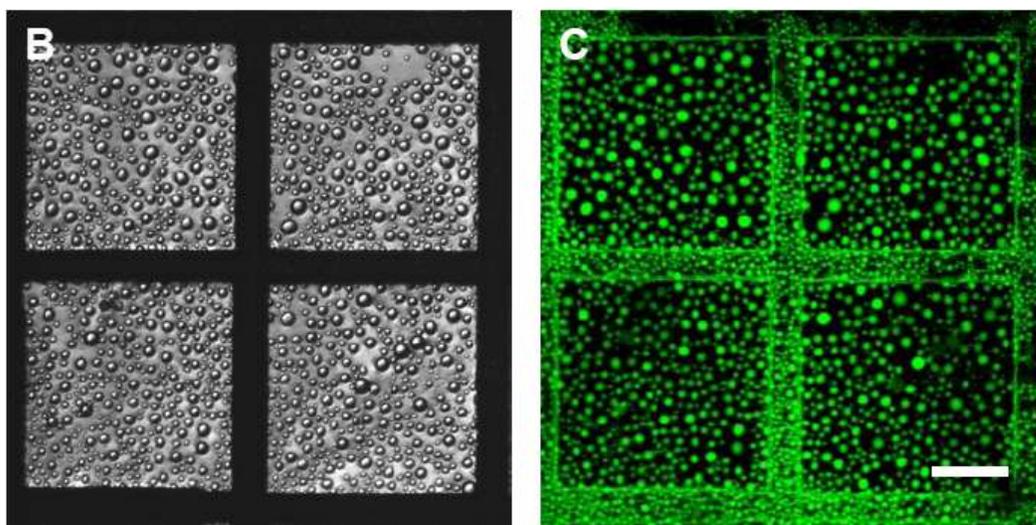
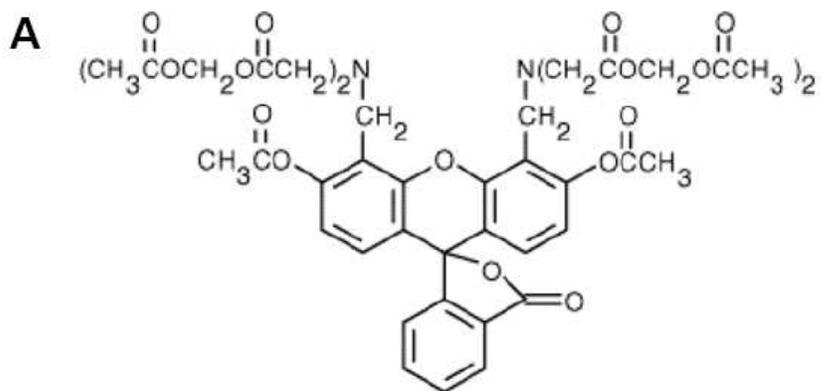


Figure S1. A) The chemical structure of calcein AM. B) and C) Images of nematic 5CB hosted in gold grids supported on OTS-treated glass slides in contact with water for 3 days and then 2  $\mu\text{M}$  calcein AM for one day. B) is a bright field image and C) is fluorescence image. The scale bar shown in C corresponds to 100  $\mu\text{m}$ .

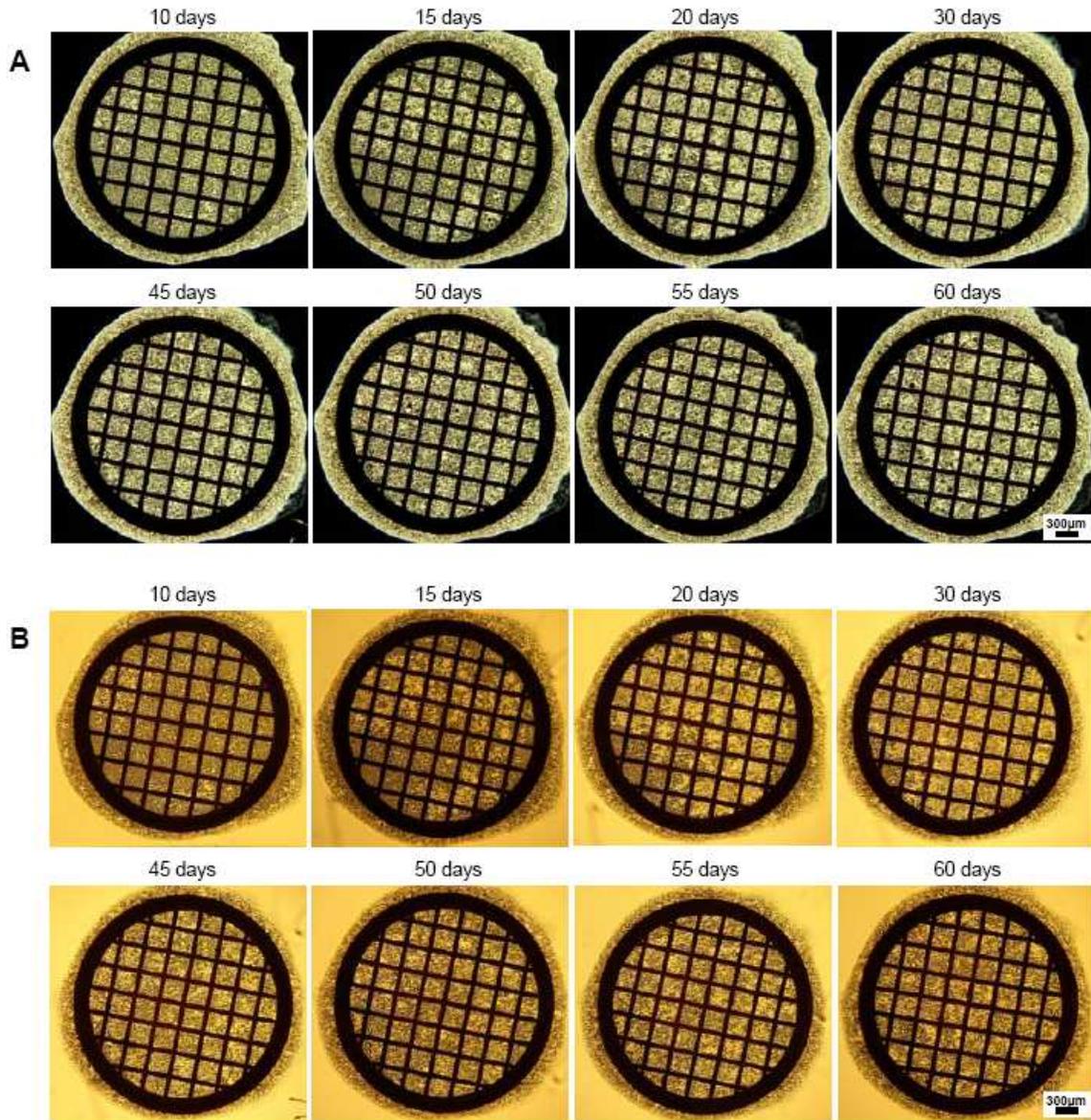


Figure S2. Optical micrographs of nematic 5CB hosted in gold grids supported on OTS-treated glass in contact with water: A) polarized light micrographs (crossed polars) and B) bright field images.

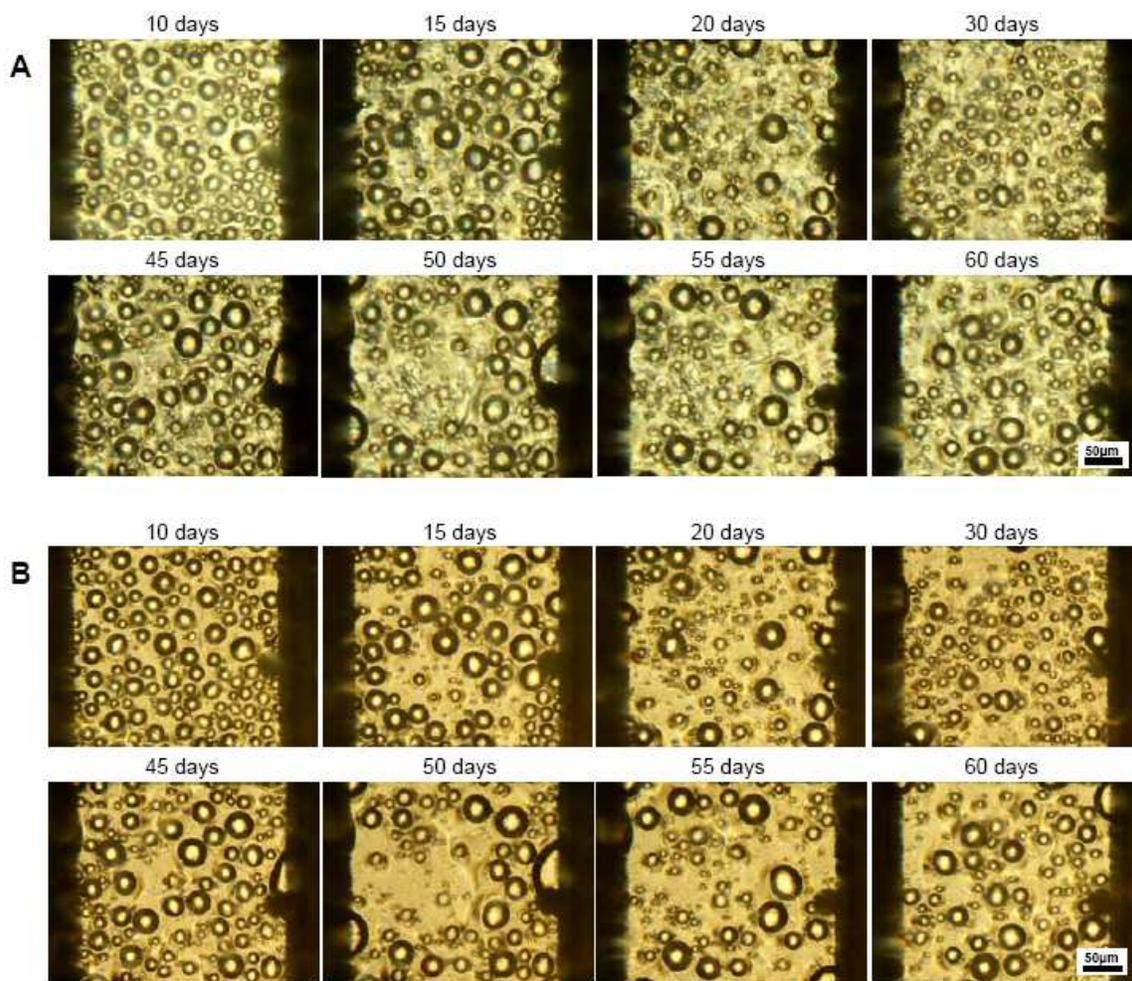


Figure S3. Optical micrographs of nematic 5CB hosted in gold grids supported on OTS-treated glass in contact with water: A) polarized light images (crossed polars) and (B) bright field images.

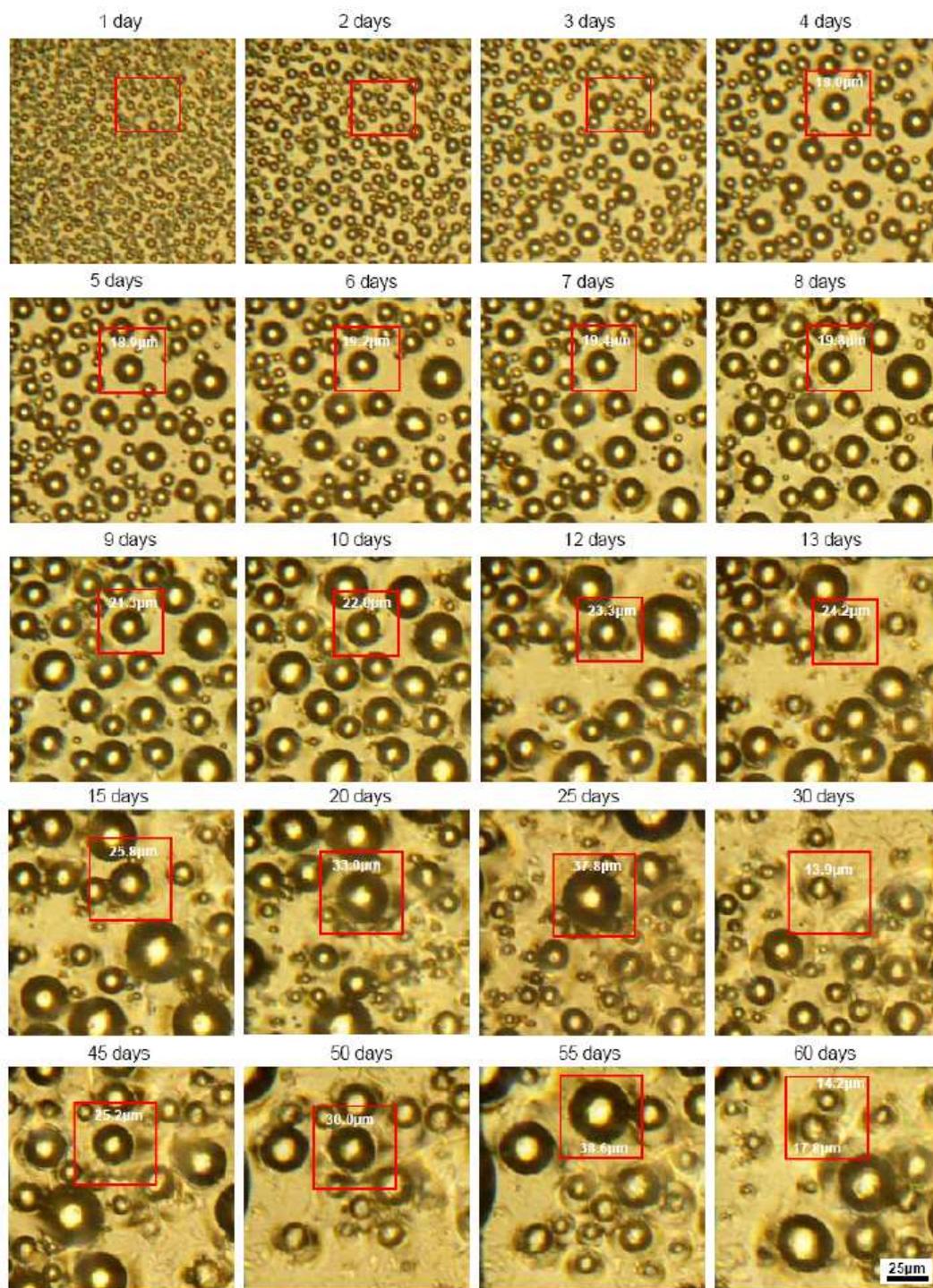


Figure S4. Optical micrographs of nematic 5CB hosted in gold grids supported on OTS-treated glass substrates in contact with water. The red box tracks the change in size of a particular droplet, the size of which is indicated in the image.

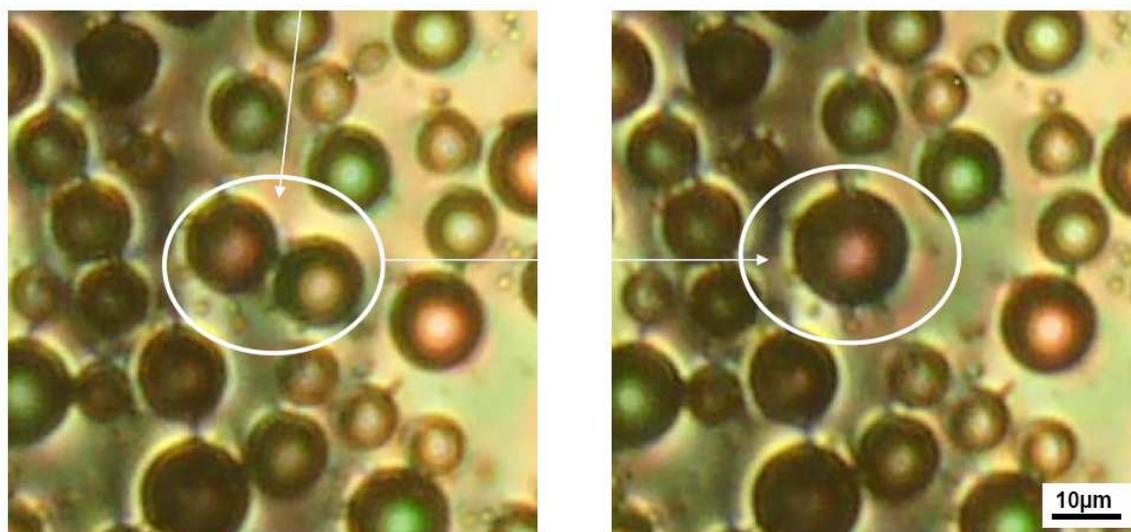
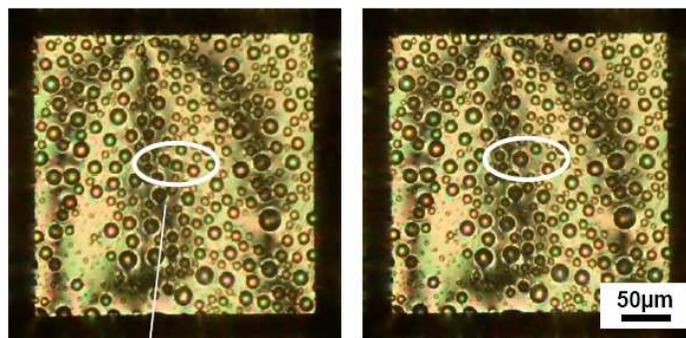


Figure S5. Polarized light micrographs (crossed polars) showing the coalescence of two water droplets (circled by white line) in nematic 5CB. The 5CB was hosted in a gold grid supported on OTS-treated glass.

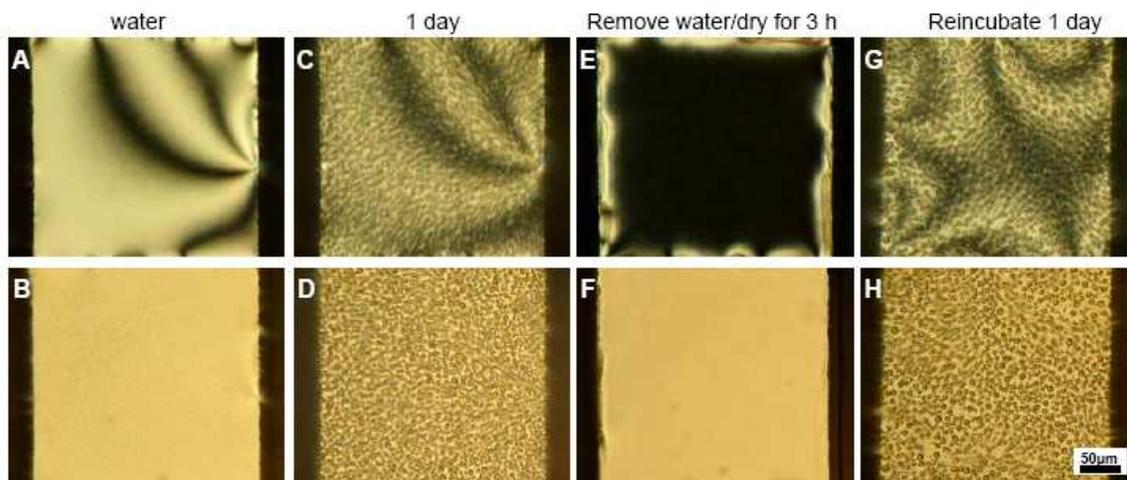


Figure S6. Optical micrographs of nematic 5CB supported on OTS-treated glass incubated in water for (A, B) 10 min and (C, D) one day, respectively. Optical micrographs of nematic 5CB on OTS-treated glass incubated in water for one day, then (E, F) removed from water and contacted with air for 3 h; (G, H) re-immersed in water for one day. The top row of images were obtained using polarized light (crossed polars) and the bottom row of images were obtained using bright field imaging.

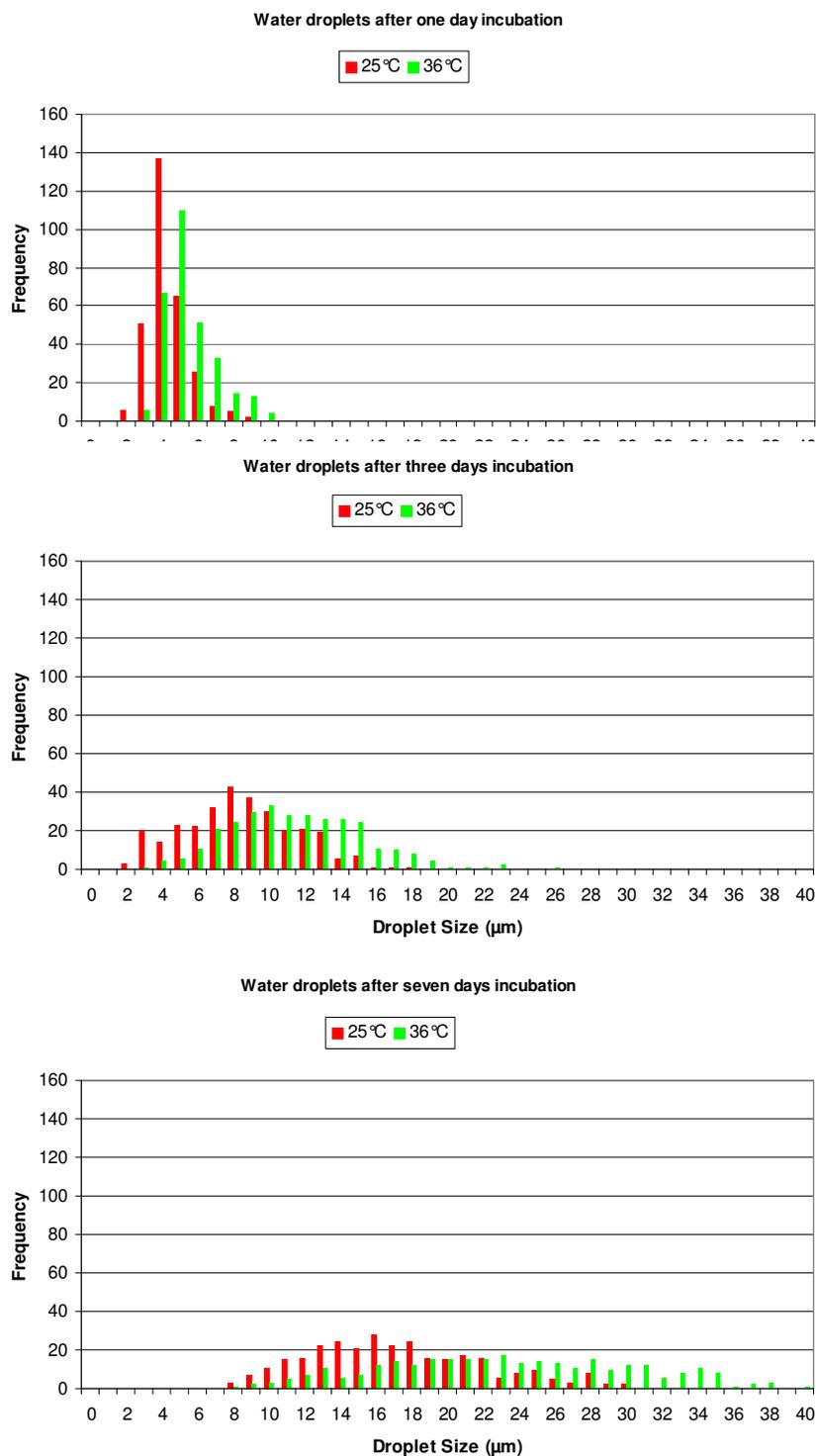


Figure S7. Evolution of sizes of water droplets in 5CB supported on OTS-treated glass after seven days incubation at 25 °C (red column) and 36 °C (green column) in water.

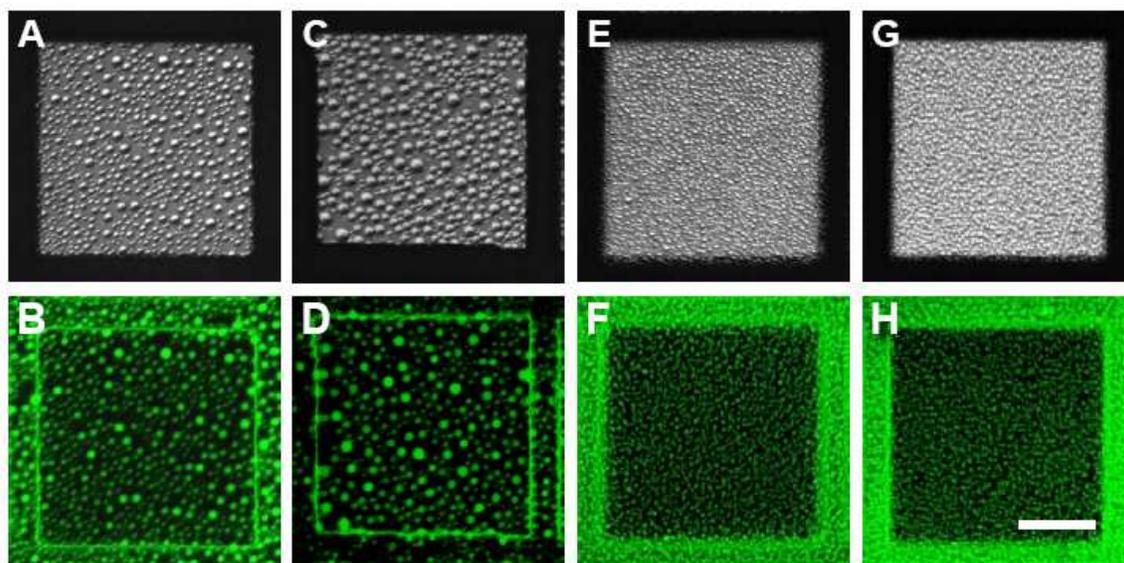


Figure S8. Optical images of (A, B) isotropic silicone oil; (C, D) olive oil; (E, F) corn oil and (G, H) vegetable oil hosted in gold grids supported on OTS-treated glass, in contact with water for 6 days and then 2  $\mu\text{M}$  calcein AM for one day. The top images are bright field micrographs and the bottom images are fluorescence micrographs. The scale bar corresponds to 100  $\mu\text{m}$ .

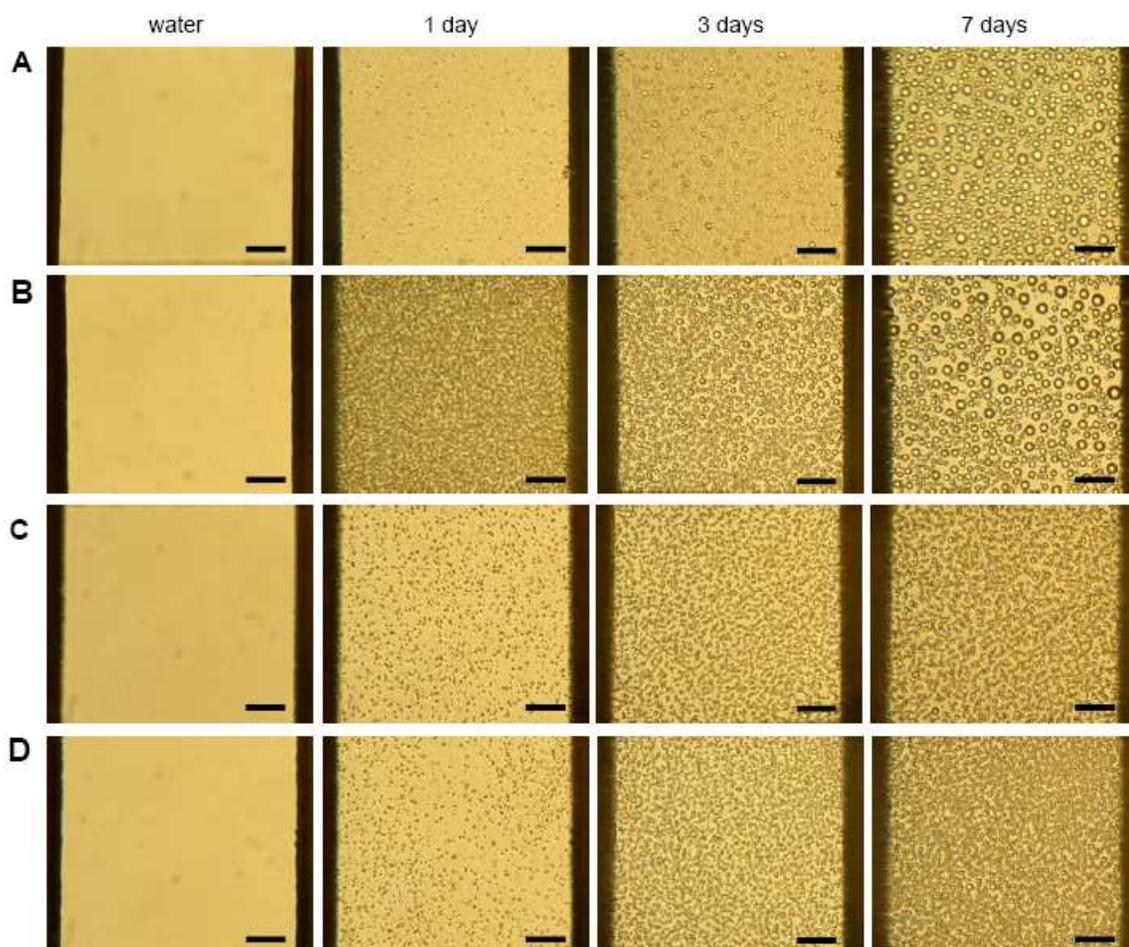
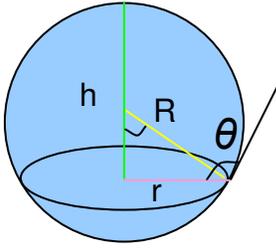


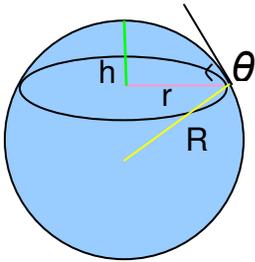
Figure S9. Optical micrographs of isotropic (A) silicone oil; (B) olive oil; (C) corn oil and (D) vegetable oil hosted in gold grids supported on OTS-treated glass, in contact with water for seven days. The scale bar corresponds to 50  $\mu\text{m}$ .



$$A_1 = \pi r^2 = \pi [R \sin(180^\circ - \theta)]^2 = \pi R^2 \sin^2 \theta$$

$$A_2 = 2\pi R h = 2\pi R [R + R \cos(180^\circ - \theta)] = 2\pi R^2 (1 - \cos \theta)$$

$$\boxed{\frac{A_2}{A_1} = \frac{2(1 - \cos \theta)}{\sin^2 \theta}}$$



$$A_1 = \pi r^2 = \pi R^2 \sin^2 \theta$$

$$A_2 = 2\pi R h = 2\pi R [R - R \cos \theta] = 2\pi R^2 (1 - \cos \theta)$$

$$\boxed{\frac{A_2}{A_1} = \frac{2(1 - \cos \theta)}{\sin^2 \theta}}$$

Figure S10. Derivation of the relationship between  $A_1$ ,  $A_2$  and  $\theta$ , where  $A_1$  refers to the contact area between a water droplet and a solid substrate,  $A_2$  refers to the contact area between a water droplet and oil phase. The contact angle  $\theta$  is either larger or smaller than  $90^\circ$  as shown in the top and bottom illustrations, respectively.

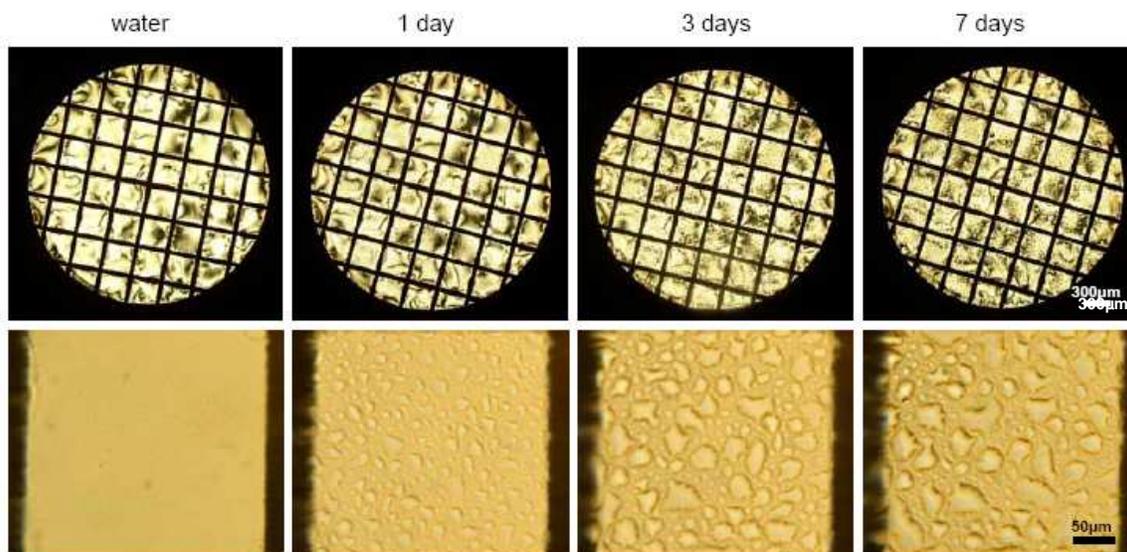


Figure S11. Optical micrographs of nematic 5CB hosted in a gold grid supported on a glass slide (not treated with OTS) in contact with water. The top images are polarized light micrographs and the bottom images are bright field micrographs.

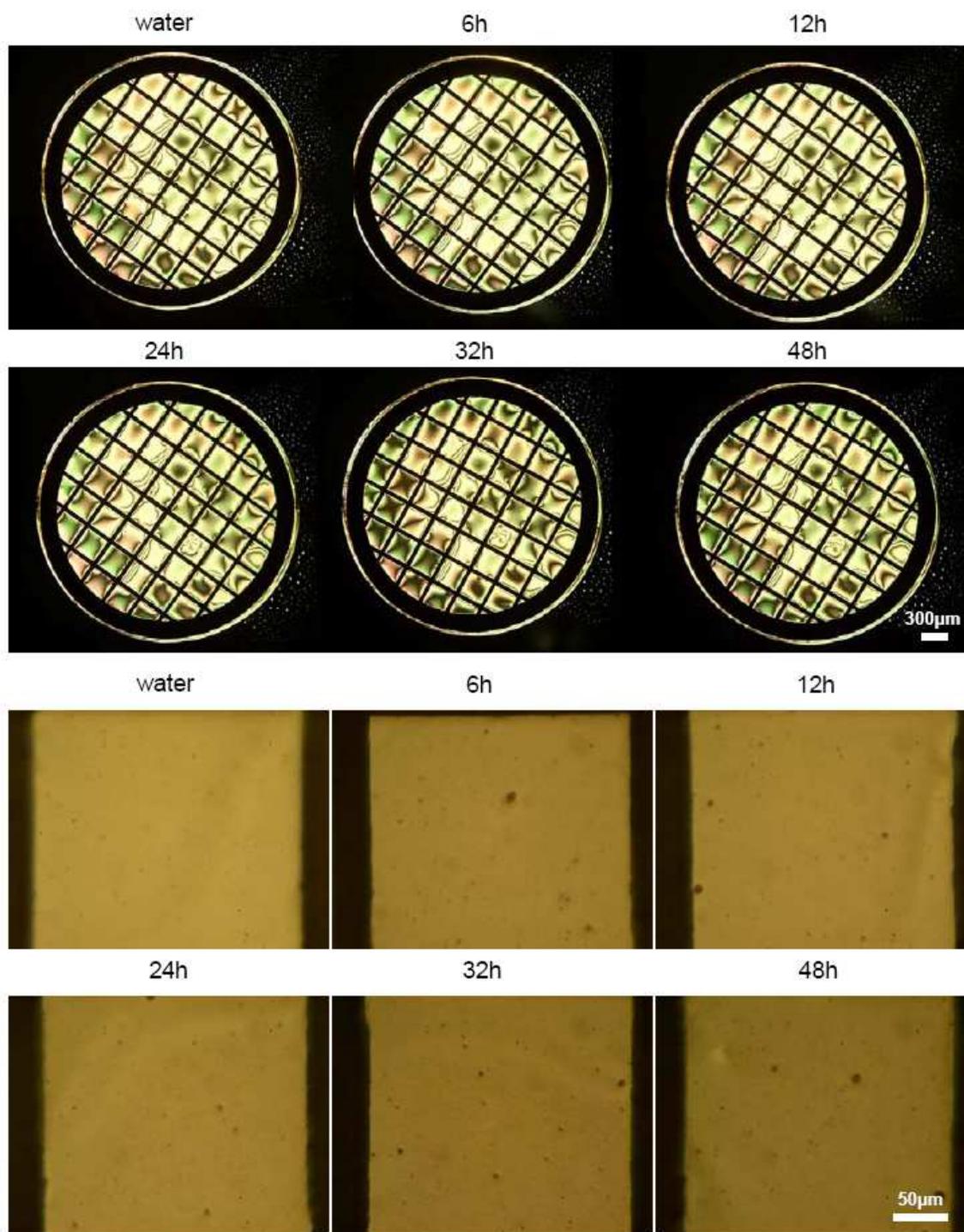


Figure S12. Optical micrographs of nematic 5CB hosted in gold grids supported on mixed monolayers formed from  $C_{10}SH$  and  $C_{16}SH$  on a gold film, in contact with water. The top images are polarized light micrographs and the bottom images are bright field micrographs.

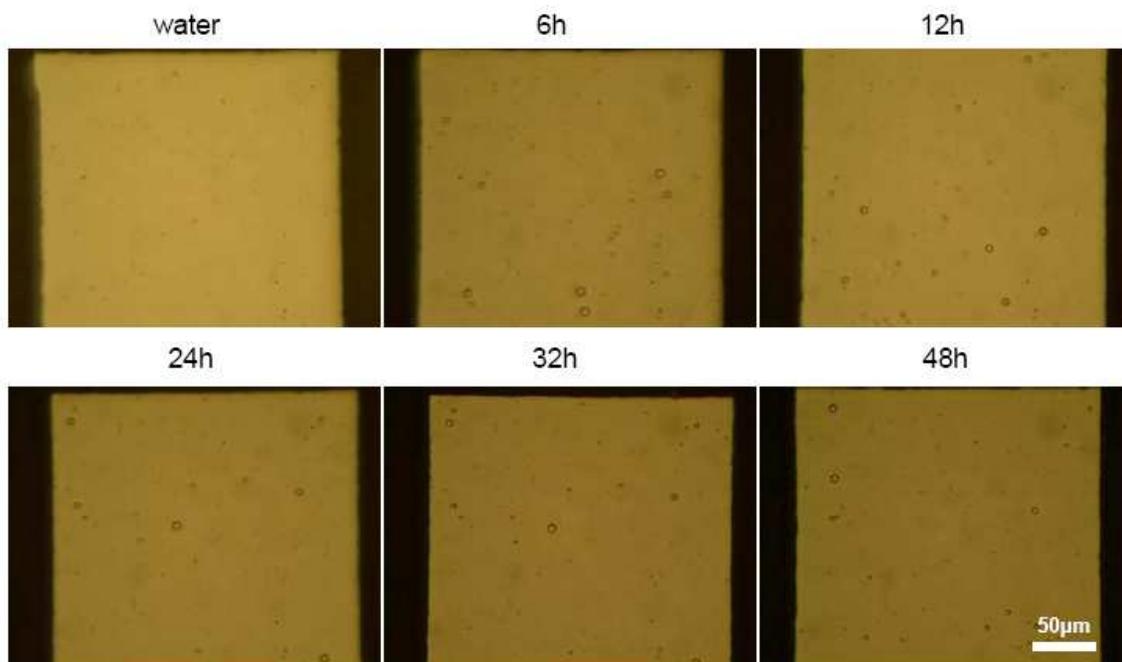


Figure S13. Optical micrographs of isotropic silicone oil hosted in gold grids supported on mixed monolayers formed from  $C_{10}SH$  and  $C_{16}SH$  on a gold film, in contact with water.

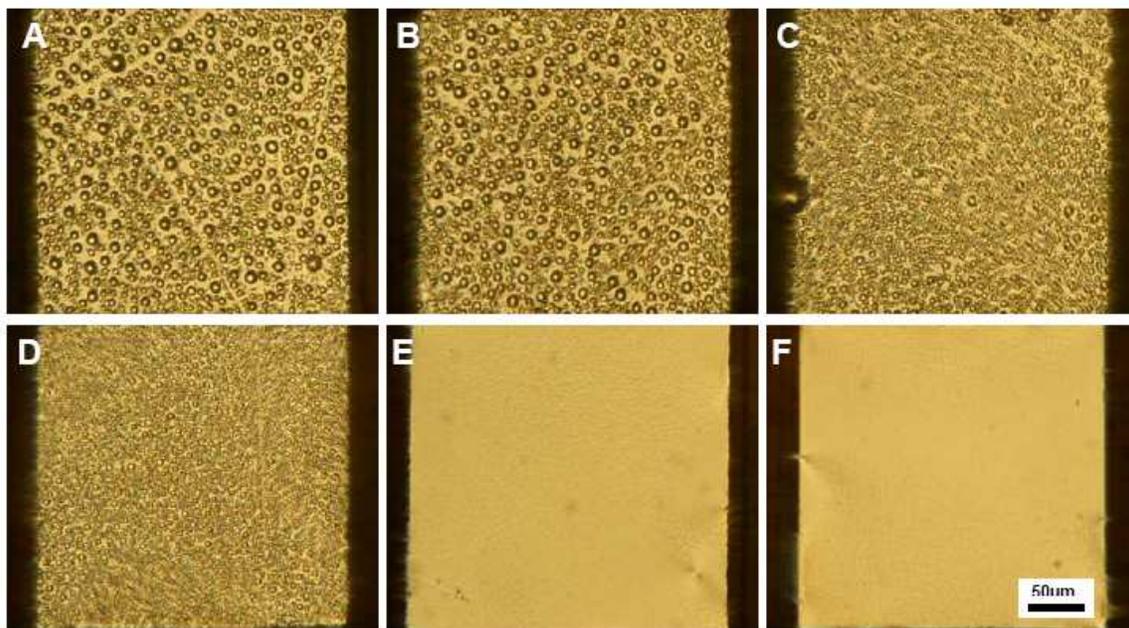


Figure S14. Optical micrographs of nematic 5CB hosted in gold grids supported on OTS-treated glass slides in contact with aqueous NaCl for three days. The concentration of NaCl in the aqueous phase is (A) 0.01 mM; (B) 0.1 mM; (C) 1 mM; (D) 10 mM; (E) 100 mM or (F) 200 mM.

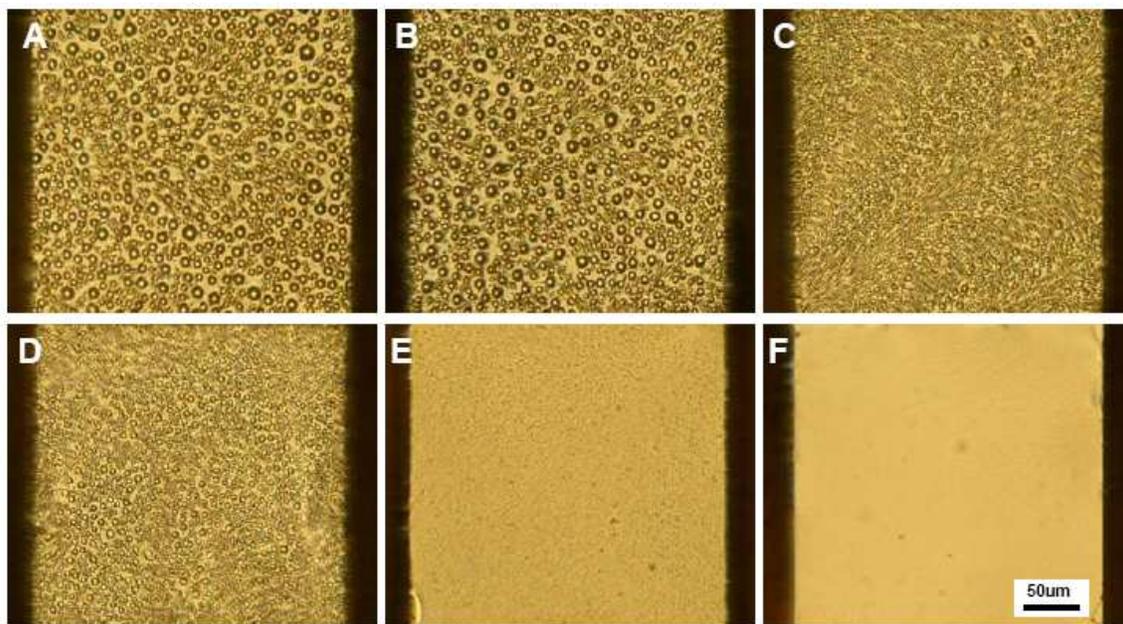


Figure S15. Optical micrographs of nematic 5CB hosted in gold grids supported on OTS-treated glass slides in contact with aqueous sucrose for three days. The concentration of the sucrose in the water was (A) 0.01 mM; (B) 0.1 mM; (C) 1 mM; (D) 10 mM; (E) 100 mM or (F) 300 mM.