Electronic Supplementary Information for

Controlled nucleation in evaporative crystallization using confined-vapor driven solutal Marangoni effect

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figure S1.pdf

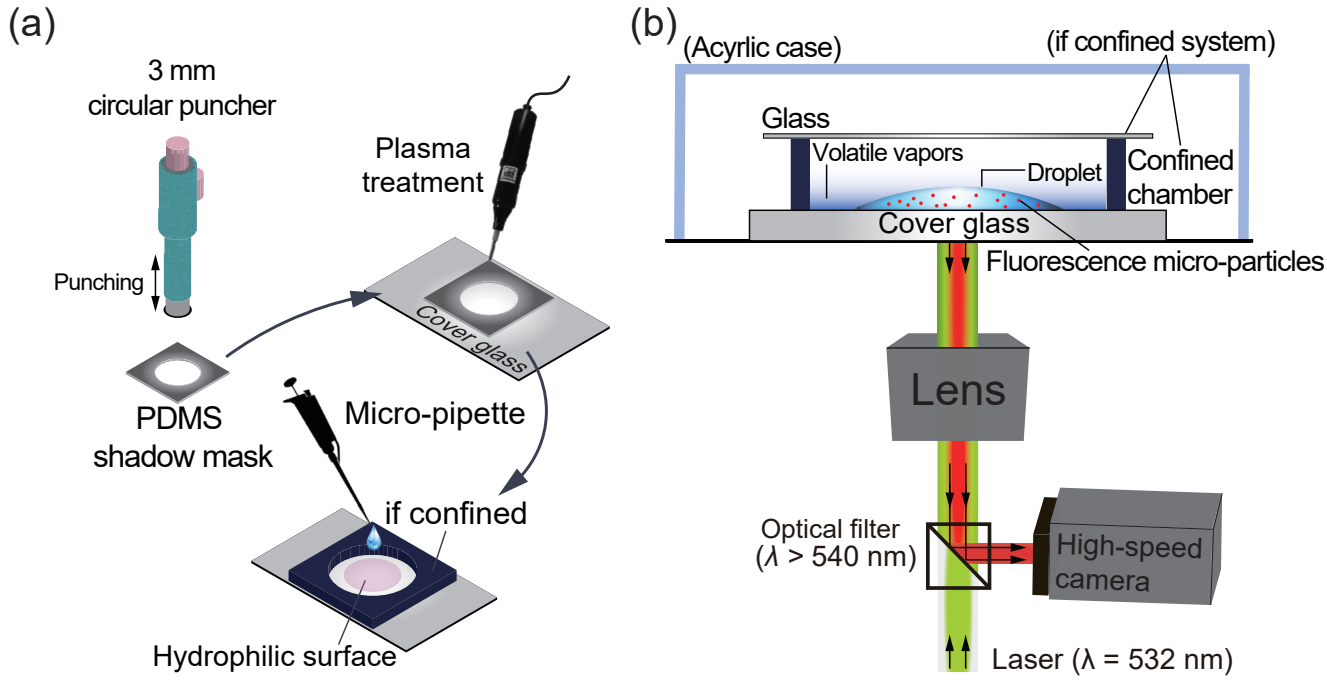

Fig. S1 Experimental setup. (a) Preparation of a liquid droplet with a constant 3 mm diameter on a cover glass. The wetting area is treated with a plasma to make locally hydrophilic. (b) Experimental setup for microparticle image velocimetry (PIV) measurements

Table S1 Critical Micelle Concentration (CMC) of CTAB, TTAB, or DTAB in an ethanol-water mixture at $T \approx 25$ °C

Surfactants	Chemical formula	CMC (mM, in ethanol : water = 30 : 70 vol.%)
C_{16}TAB (CTAB)[1]	$CH_3(CH_2)_{15}N(Br)(CH_3)_3$	2.9[1]
C_{14}TAB (TTAB)[1]	$CH_3(CH_2)_{13}N(Br)(CH_3)_3$	5.4[1]
C_{12}TAB (DTAB)[1]	$CH_3(CH_2)_{11}N(Br)(CH_3)_3$	37.4[2]

[1] $\geq$ 99% purity, Sigma-Aldrich, U.S.A.

References

[1] S. K. Shah, S. K. Chatterjee and A. Bhattarai, *Journal of Molecular Liquids*, 2016, **222**, 906-914.

[2] S. Aslanzadeh and A. Yousefi, *Journal of Surfactants and Detergents*, 2014, **17**, 709-716.

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[†] Electronic Supplementary Information (ESI) available: [details of any supplementary information available should be included here]. See DOI: 10.1039/cXsm00000x/

figure S2.pdf

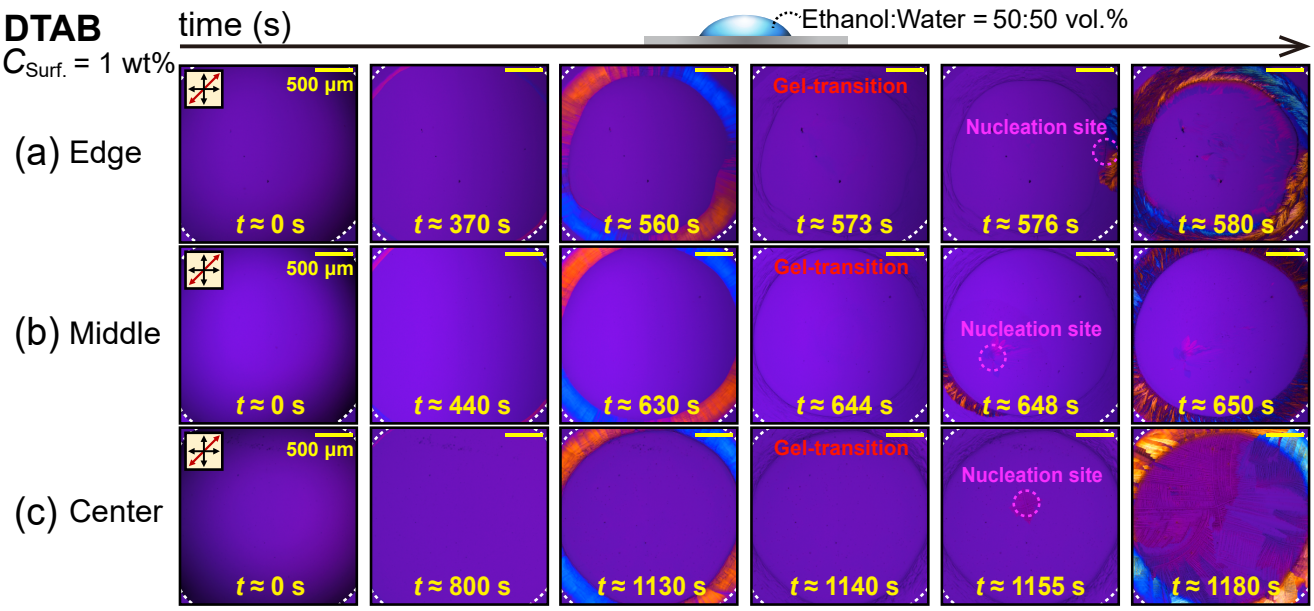

Fig. S2 Evaporation of DTAB-containing binary mixture droplets in an open space. Non-controllable nucleation sites (purple dashed-circles) in the vicinity of the (a) edge, (b) middle between the edge and center, and (c) center of 1 wt% DTAB in ethanol and water mixture (50:50 vol.%) droplets. Here, we compared all sequential POM images for the entire evaporation process of the DTAB droplets. Initial contact lines are displayed by white dashed-lines. All scale bars are 500 μm.

figure S3.pdf

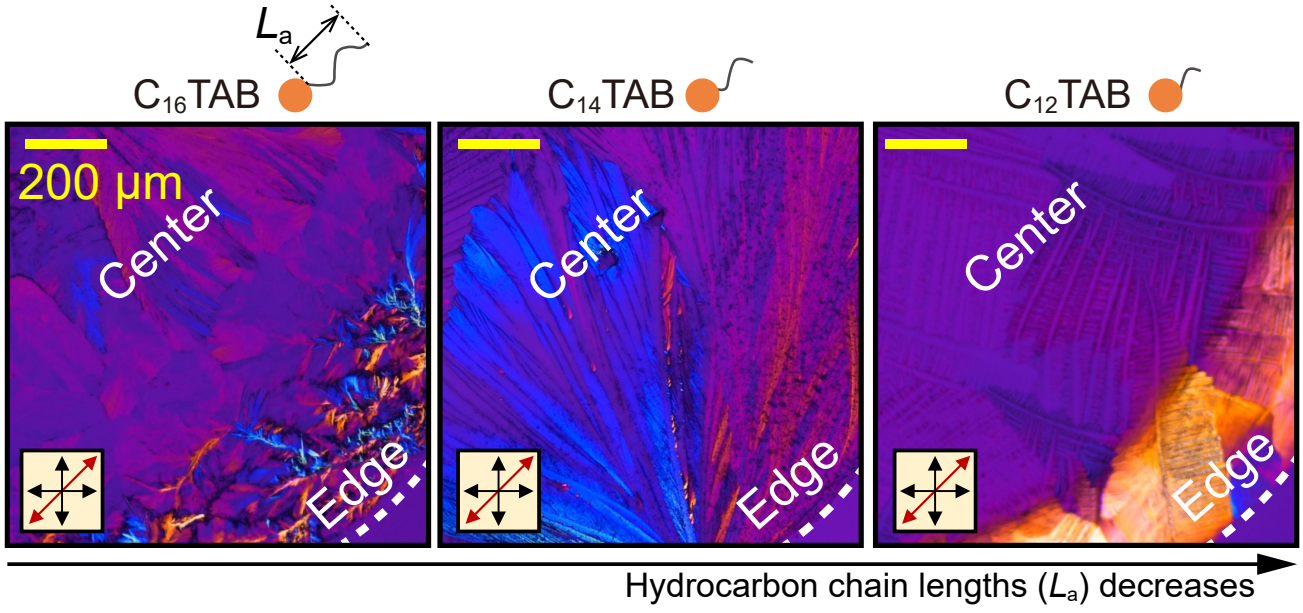

Fig. S3 Comparison of crystalline morphologies depending on hydrocarbon chain length L_a. We compared the crystal growth results of three different cationic surfactants [here, CTAB (C_{16}TAB), TTAB (C_{14}TAB), and DTAB (C_{12}TAB)] in the vicinity of the droplet contact line (the white dashed-lines). All crystalline patterns were observed after the surfactant (1 wt%)-containing ethanol-water mixture (50:50 vol.%) droplets totally evaporated on an open substrate. All scale bars are 200 μm.

figure S4.pdf

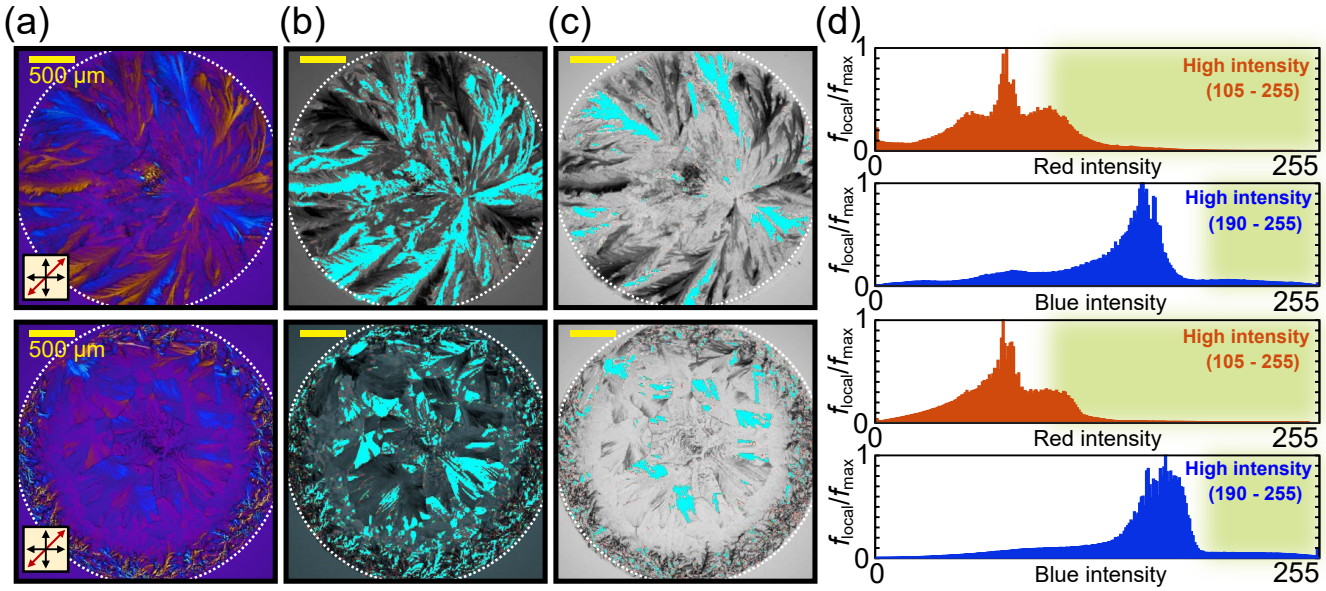

Fig. S4 Process of detecting a yellowish and bluish area of (a) POM images produced after 1 wt% CTAB-containing ethanol and water (50:50 vol.%) droplets evaporated in a confined (the first row) or an open system (the second row). We distinguished (b) Yellow- and (c) blue-colored crystalline regions exhibiting a relatively high light intensity highlighted as cyan areas in (b) and (c) [here, in the range of 105-255 (red) and 190-255 (blue) in (d), respectively] as shown in greenish areas of (d) red-and-blue histograms with a relative frequency value $f_{\mathrm{local}}/f_{\mathrm{max}}$. All scale bars are 500 μm.