

Emergent collective motion of self-propelled condensate droplets

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List of supporting videos

Video S1. Interdroplet attraction between sub-millimetric droplets (original speed) on cooled micropillar surface lubricated with fluorinated oil (Krytox GPL 100, $\eta = 53$ mPa.s at $T = 4^\circ\text{C}$).

Video S2. Serpentine motion of droplet condensates on cooled micropillar surface lubricated with fluorinated oil (Krytox GPL 100, $\eta = 53$ mPa.s at $T = 4^\circ\text{C}$). Scale bar is 2 mm.

Video S3. Droplet motion tracked automatically using Circle Hough Transform with droplet radii (in pixels) indicated next to outline.

Video S4. Serpentine droplets continually seek areas with *locally* higher ϕ_l . Video is sped up 50 $\times$, and the field of view is 22.5 mm $\times$ 18 mm.

Video S5. Zoomed-in view of droplet condensates on cooled micropillar surface lubricated with fluorinated oil (Krytox GPL 101, $\eta = 73$ mPa.s at $T = 4^\circ\text{C}$). Field of view 6.7 mm $\times$ 5.1 mm.

Video S6. Collective motion of hot vapour condensates on nanostructured surface lubricated with fluorinated oil (Krytox GPL 102, $\eta = 15$ mPa.s at $T = 60^\circ\text{C}$). Scale bar is 1 cm. Serpentine and circular motions co-exist.

Video S7. Collective motion of droplet condensates on cooled micropillar surface lubricated with fluorinated oil (Krytox GPL 100, $\eta = 53$ mPa.s at $T = 4^\circ\text{C}$). Scale bar is 4 mm. Serpentine and circular motions co-exist.

Video S8. Collective motion of hot vapour condensates (original speed) on nanostructured surface lubricated with silicone oil ($\eta = 2$ mPa.s at $T = 60^\circ\text{C}$). Serpentine and circular motions co-exist.

Video S9. After more than 4 hours of cooling, the surface is now depleted of lubricated with no lubricant overlayer. There is no more droplet self-propulsion. Note that Video S9 is taken at a later timepoint from Video S5.

Materials and Methods

Table S1: Viscosity (in mPa.s) of different lubricants at different temperatures.

Temperature ($^\circ\text{C}$)	Silicone oil	Krytox GPL			
		100	101	102	103
4	–	53	73	173	–
25	5	21	–	55	144
40	3	12	19	29	64
60	2	6.6	–	15	34

Material and sample preparation. Fluorinated oils (Krytox GPL 100, 101, 102, 103) were purchased from Chemours company, while silicone oil was purchased from Sigma Aldrich. We measured their viscosities η at different temperatures using a rheometer/viscometer as summarized in Table S1. Deionized (DI) water with a resistivity of $18.2 \text{ M}\Omega \text{ cm}^{-1}$ was obtained from a Milli-Q water purification system (Millipore, Bedford, MA, USA). The surface tension γ of water at room temperature is 72 mN m^{-1} . At the different experimental $T = 4$ to 60°C , γ varies by about 10%. This is in sharp contrast to η which changes by one order of magnitude due to the temperature change.

Cooling experiments. We use a peltier system to cool the surface to $T = 4^\circ\text{C}$. For cooling experiments, we used a micropillar silicon substrate (hexagonal array with diameter $d = 2 \text{ }\mu\text{m}$, pitch $p = 10 \text{ }\mu\text{m}$, and height $h_p = 9 \text{ }\mu\text{m}$) made using conventional photolithography

(Bosch process) and hydrophobized by radio-frequency plasma sputtering of 100 nm thick fluoropolymer coating (C_4F_8 gas for 15 min).¹ Different lubricants (typically fluorinated oils) was then spincoated onto the surface, and its thickness measured using white-light interferometry as explained in our previous paper.² Typically, we start with an overlayer thickness of $h_o = 5 \text{ }\mu\text{m}$. We used an infrared (IR) camera FLIR SC5000 to image condensate droplets for Figs. 2(b) and 5(a) in the main text. Note that the different grayscale values are not due to temperature variations, but rather different material emissivity when water condenses on the surface. The underlying silicon wafer has a low emissivity and will reflect IR from the surrounding into the camera (hence appears white when not covered by water). In contrast, water has a high emissivity and hence appears black on the camera (since it is cold). For zoomed-in view in Figs. 1(b) and 3, we used a microscope objective ($5\times$ Mitutoyo Plan Apochromat) attached to a high-resolution 4K camera (Panasonic GH5).

Heating experiments. We heated 60 mL of water in a 150 mL beaker to a temperature of 55°C . We then placed the lubricated surface (upside down) on top of the beaker, with the lubricant layer facing the hot vapour. The temperature of the substrate stayed relatively constant at $T = 40^\circ\text{C}$ throughout experiment, as measured using a thermocouple. For all heating experiments, we used nanostructured glass surface lubricated with either silicone or fluorinated oils. To create the nanotextures, we spray coated clean glass slides ($5 \text{ cm} \times 7.5 \text{ cm}$) with a layer of hydrophobic nanoparticles (Glaco Mirror Coat Zero, Soft 99 Co.) and placed them vertically to dry for one hour before spincoating with the lubricant oil. We applied an initial $h_o = 15 \text{ }\mu\text{m}$ for the nanotextured surface. For all heating experiments, we imaged the condensate droplets using a DSLR camera and a macro-lens (Canon MPE 65 mm 1-5 $\times$ Macro Photo Lens).

The phenomenon observed here is not sensitive to the exact lubricant film thickness. Practically, we found that an *initial* $h_o = 5 \text{ }\mu\text{m}$ for the micro-pillar and $15 \text{ }\mu\text{m}$ for the nanotextured surfaces are ideal to jumpstart the serpentine motion. Note, however, that once

the serpentine motions start, there will be significant temporal and spatial variations in h_o which drive the complex collective motion described in this paper, including the transition from serpentine to circular motions. In all our experiments (heating and cooling), the ambient room temperature was kept at 22°C and the room humidity varied between 40 and 60%.

Attraction between similarly-sized droplets

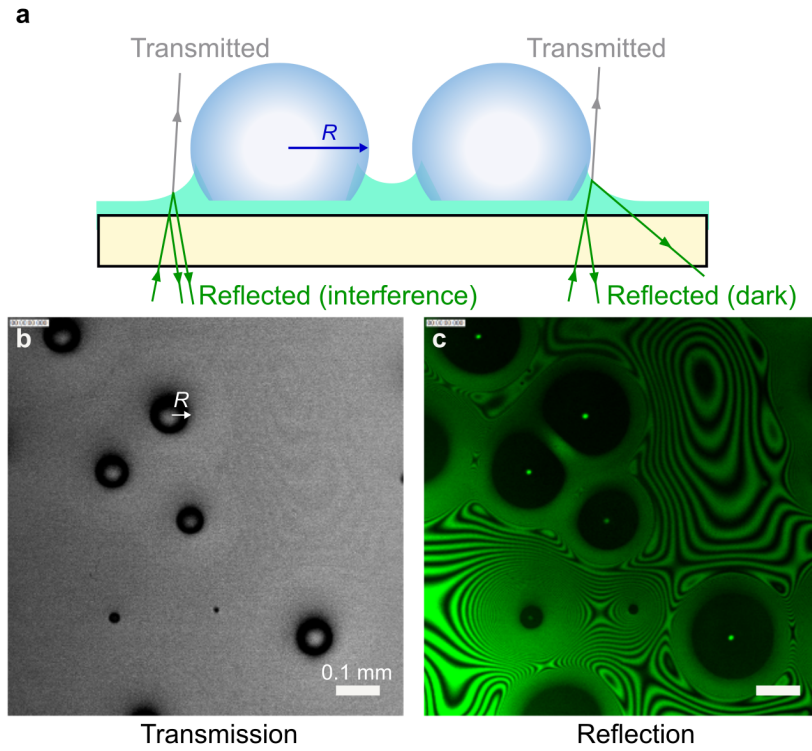


FIG. S1: **Attraction between similarly-sized droplets:** (a) Schematic showing how the reflected and transmitted signals are collected when shining monochromatic light from under the surface. (b)–(c) Transmission and reflection signals, respectively. Scale bars are 0.1 mm.

We shone monochromatic light with wavelength $\lambda = 561$ nm from under the surface and captured both the transmitted and reflected signals simultaneously (from above and below, respectively; see schematic in Fig. S1(a)). The technique described here is known as reflection interference contrast microscopy and has been reported extensively by our group

and others.²⁻⁴

The droplet radius R can be measured accurately from the transmitted signal [Fig. S1(b)], while the distortion to the lubricant layer is best visualized from the reflected signal [Fig. S1(c)]. In the presence of a thin and relatively *flat* film, light reflected off the solid/lubricant and lubricant/air interfaces interfere with one another constructively or destructively to give bright or dark fringes, respectively. There is a change in lubricant thickness $\Delta h = \lambda/4n_o \approx 100$ nm when moving from one bright fringe to a neighbouring dark fringe, where $n_o = 1.3$ is the refractive index of the lubricant.

The dark regions in Fig. S1(c) correspond to the wetting ridge around each droplet: the distortion to the lubricant film results in a gradient so steep that the light reflected off the lubricant/air interface cannot reach the camera, hence the reduced light intensity [See Fig. S1(a)]. In the middle of Fig. S1(c), we see three neighbouring and similarly-sized droplets attracting one another; their wetting ridges overlap and the resulting menisci are non-axisymmetric, resulting in a net attractive force.

Note that in this experiment, we chose a relatively viscous lubricant (K103, $\eta = 144$ mPa.s at $T = 25^\circ$) to slow down the droplet dynamics and hence better capture the light signals.

Attraction between polydisperse droplets

The schematic and images in Fig. S2 demonstrate the capillary attraction that exists between one large millimetric droplet and its neighbouring (and much smaller) droplets. The time series in Fig. S2(b) show one small droplet of radius $r = 30 \mu\text{m}$ (circled) is attracted towards a droplet with a much larger $R = 1.2 \text{ mm}$ moving at a speed $u \approx 0.1 \text{ mm s}^{-1}$. The overlap in the droplet's wetting ridges is again evident from the reflection signal in Fig. S2(b). The serpentine motion in Fig. 2 of the main text is similarly driven by capillary attraction as depicted here, except the attraction is between one large millimetric droplet and multiple

(not just one) sub-millimetric droplets.

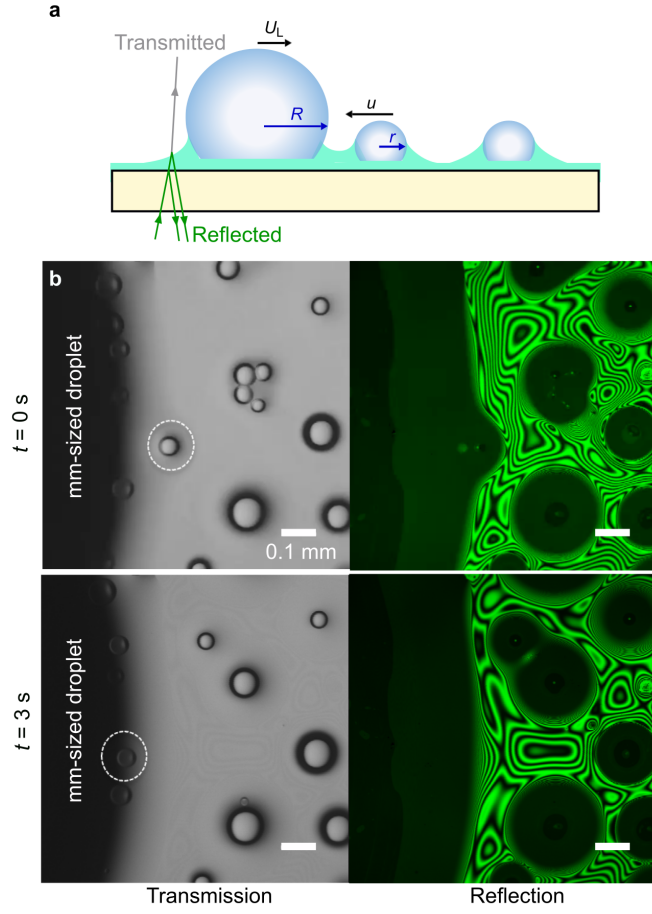


FIG. S2: **Attraction between differently-sized droplets:** (a) Schematic and (b) images illustrating the capillary attraction between one large droplet and its neighbouring small droplets. Time series in (b) show how one small droplet (circled) moves towards a much larger millimetric-sized droplet.

Note that in this experiment, we chose a relatively viscous lubricant (K103, $\eta = 144$ mPa.s at $T = 25^\circ$) to slow down the droplet dynamics and hence better capture the light signals. In the experiments presented in the main manuscript, the lubricants used were significantly less viscous and we expect much faster droplet dynamics.

Energy balance for a self-propelled droplet

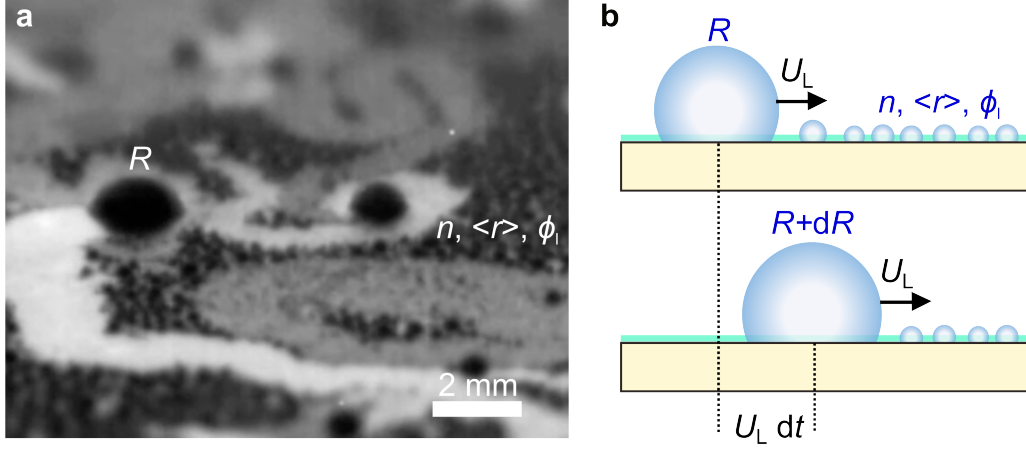


FIG. S3: (a) Sideview image and (b) schematic for a millimetric self-propelled droplet of radius R moving at speed U_L among a sea of smaller droplets with mean radius $\langle r \rangle$ and number density (per unit area) n . The *local* surface coverage ϕ_l for the smaller droplets is equivalent to $n\pi\langle r^2 \rangle$.

Using energy balance, we will derive an expression for the expected speed U_L for a self-propelled droplet in a serpentine motion. Consider a millimetric-sized droplet of radius R (and volume V) moving at a speed U_L among a sea of smaller droplets with mean radius $\langle r \rangle$ and number density (per unit area) n . After a short time dt , the droplet has moved by a distance $U_L dt$ and swept over an area $2RU_L dt$ [Fig. S3]. We therefore expect the increase in droplet's volume to be

$$\begin{aligned} dV &= \frac{2}{3}n\pi\langle r^3 \rangle(2RU_L dt) \\ &= \frac{4}{3}n\pi\langle r^3 \rangle RU_L dt \end{aligned} \tag{S1}$$

Hence, we expect the growth rate in the droplet's radius to be

$$\begin{aligned} \frac{dR}{dt} &= \frac{dV}{dt} \frac{dR}{dV} \\ &= \frac{4}{3}n\pi\langle r^3 \rangle RU_L \cdot \frac{1}{4\pi R^2} \\ &= \frac{n\langle r^3 \rangle U_L}{3R} \end{aligned} \tag{S2}$$

As the large droplet gobbles up the smaller droplets, there is a decrease in the droplets' *total* surface area A_{tot} , i.e.,

$$\begin{aligned}
dA_{\text{tot}} &= 2\pi(R + dR)^2 - 2\pi R^2 - 2n\pi\langle r^2 \rangle (2RU_L dt) \\
&= 2\pi(R^2 + 2RdR) - 2\pi R^2 - 4n\pi\langle r^2 \rangle RU_L dt \\
&= 4\pi R dR - 4n\pi\langle r^2 \rangle RU_L dt
\end{aligned} \tag{S3}$$

Comparing Eqs. (S2) and (S3) gives

$$\begin{aligned}
\frac{dA_{\text{tot}}}{dt} &= 4\pi R \frac{dR}{dt} - 4n\pi\langle r^2 \rangle RU_L \\
&= 4n\pi\langle r^2 \rangle RU_L \left(\frac{\langle r^3 \rangle}{3R\langle r^2 \rangle} - 1 \right) \\
&\approx 4n\pi\langle r^2 \rangle RU_L \left(\frac{\langle r \rangle}{3R} - 1 \right) \\
&\approx -4n\pi\langle r^2 \rangle RU_L \\
&= -4RU_L\phi_l
\end{aligned} \tag{S4}$$

. Here, we approximate the droplet geometry as a hemisphere and assume that $\langle r \rangle / R \ll 1$ (reasonable assumption if we look sideview image in Fig. S3(a)), where $\phi_l = n\pi\langle r \rangle^2$ is the *local* fractional surface coverage of the smaller droplets. We can also check for the consistency of our assumption by comparing the relative magnitudes of $\pi \frac{dR}{dt}$ and $U_L\phi_l$, since we need $U_L\phi_l \gg \pi \frac{dR}{dt}$ for Eq. (S4) to be true. For Fig. 2 in the main text, $U_L\phi_l = 20 \mu\text{m s}^{-1}$ (assuming that $\phi_l = 0.2$) which is significantly larger than $\pi \frac{dR}{dt} \approx 4 \mu\text{m s}^{-1}$. For Fig. 3(a)–(b) in the main text, $U_L = 12 \mu\text{m s}^{-1}$ and $\frac{dR}{dt} = 0.1 \mu\text{m s}^{-1}$: $U_L\phi_l = 2.4 \mu\text{m s}^{-1}$ is significantly larger than $\pi \frac{dR}{dt} \approx 0.3 \mu\text{m s}^{-1}$.

The decrease in A_{tot} (hence, the negative sign in Eq. (S4)) results in a release of interfacial energy, though only a fraction of which $\alpha < 1$ is converted to the droplet's kinetic energy at

a rate of

$$\frac{dE_\gamma}{dt} = 4\alpha\phi_l R U_L \gamma \quad (\text{S5})$$

which must be balanced by viscous dissipation. Previously, we showed that the viscous force for a droplet moving on lubricated surface is given by $F_\eta = 2.6(2\pi)\gamma R \text{Ca}^{2/3} \approx 16 \gamma R \text{Ca}^{2/3}$, where $\text{Ca} = \eta U_L / \gamma$ is the capillary number, η is the lubricant's viscosity, while γ is the droplet's surface tension.² Hence, we expect

$$\begin{aligned} F_\eta U_L &= 4\alpha\phi_l R U_L \gamma \\ 16\gamma R \left(\frac{\eta U_L}{\gamma} \right)^{2/3} &= 4\alpha\phi_l R \gamma \\ U_L &= \frac{\gamma}{\eta} \left(\frac{\alpha\phi_l}{4} \right)^{3/2} \end{aligned} \quad (\text{S6})$$

The motion of the self-propelled millimetric droplet is fuelled by the coalescence of smaller droplets surrounding it. Self-avoiding walk arises because its previous path contains little to no water content (i.e., $\phi_l = 0$ and corresponding to white regions in Fig. S3) and no self-propulsion is possible as predicted by Eq. (S6). Self-similarity occurs since U_L has no dependence on droplet radius.

Changes in ϕ can be *qualitatively* deduced from Figs. S3 and S4. Droplets appear dark on the image; hence the darker the area, the higher the ϕ_l value is. This explains why the millimetric-sized droplet in Fig. S4 moves much faster in region A compared to B with measured $U_L = 0.1$ and 0.05 mm s^{-1} , respectively, consistent with predictions from Eq. (S6).

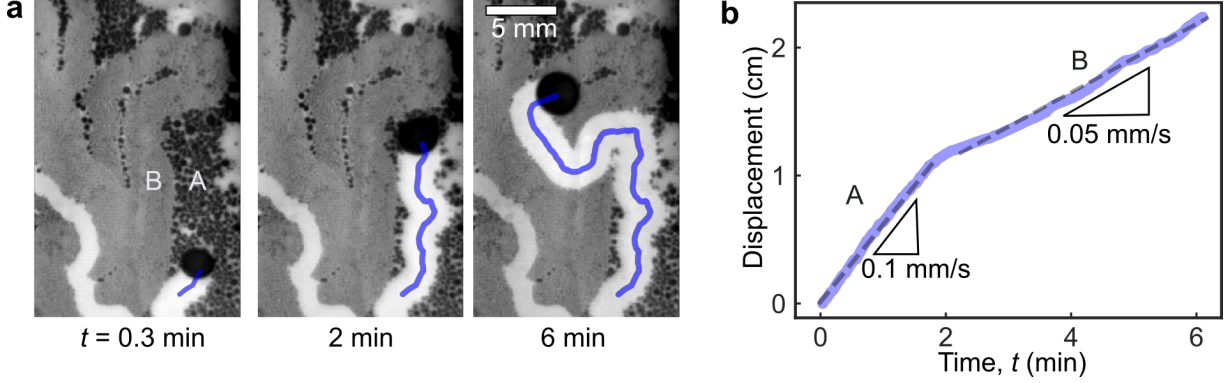


FIG. S4: (a) Serpentine motion of a droplet through two different regions A and B. A has a higher water content ϕ_l than B and hence appears darker. (b) Droplet is moving faster in A than in B consistent with predictions in Eq. (S6).

Visco-capillary vs. inertial-capillary effects

While the energy balance arguments outlined in the previous section capture the essential physics behind the droplet self-propulsion, they ignore the details of the process.

We can divide the self-propulsion into two distinct stages which cycle right after one another [Fig. S5]. In the first stage dominated by visco-capillary effects [Fig. S5(a)], the large droplet of radius R experiences capillary attraction F_γ driven by overlapping menisci with the neighbouring small droplets with distances l apart and surface area coverage ϕ_l . The resulting propulsion speed U_1 arises from a balance between F_γ and the viscous force acting on the droplet F_η . The exact forms of F_γ and F_η are difficult to model, especially when the large droplet is close to the small droplets. We expect U_1 to vary non-linearly with inter-droplet distance and hence time. In the previous section and our main manuscript, we have essentially simplified this stage by a coarse-graining approach. We assume a constant U_L which can be deduced by energy balance arguments. Since at this stage, the droplet traversed a distance of $s_1 \sim l \sim 50 \mu\text{m}$ with a typical speed of $U_L \sim 0.1 \text{ mm s}^{-1}$, the relevant timescale is $\tau_1 = l/U_L \approx 0.5 \text{ s}$. Experimentally, we found that the droplet moves with relatively constant velocity when the time-scale is larger than τ_1 consistent with our coarse-graining approach. See for example Fig. 2(c) in the main manuscript.

The second stage is dominated by inertial-capillary effects when the small droplets are

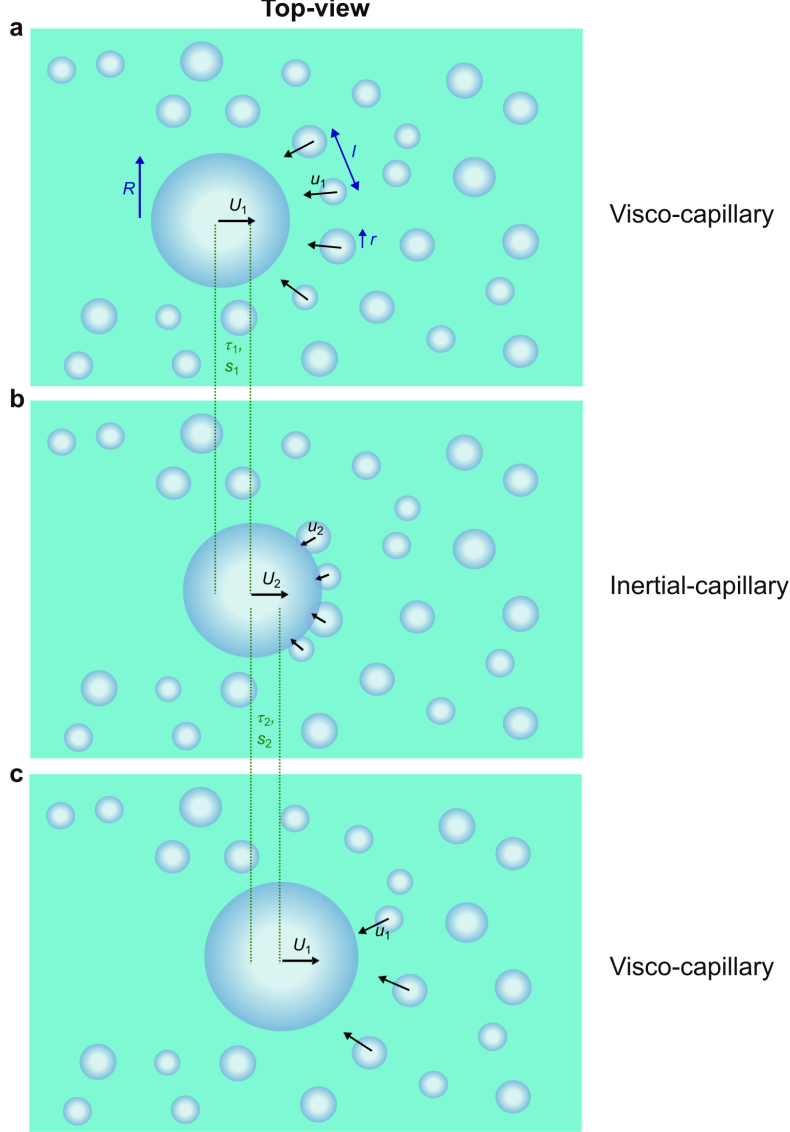


FIG. S5: The droplet self-propulsion process cycle between two distinct stages: (a) visco-capillary and (b) inertial-capillary stages. (c) Visco-capillary stage that follows right after the inertial-capillary stage.

in contact with the large droplet [Fig. S5(b)]. The small droplets will coalesce into the big droplet within the timescale $\tau_2 \sim (\rho r^3 / \gamma)^{1/2}$ and a typical velocity $u_2 \sim r / \tau_2 = (\gamma / \rho r)^{1/2}$. For typical $r \sim 20 \text{ } \mu\text{m}$, $\tau_2 \sim 10 \text{ } \mu\text{s}$ and $u_2 \sim 2 \text{ m s}^{-1}$. We can deduce the propulsion speed

of the large droplet U_2 by conservation of linear momentum, i.e.,

$$\begin{aligned}
R^3 U_2 &\sim (R/l) r^3 u_2 \\
U_2 &\sim \frac{r^3}{R^2 l} u_2 \\
&= \left(\frac{\phi_l}{\pi}\right)^{1/2} \left(\frac{r}{R}\right)^2 u_2
\end{aligned} \tag{S7}$$

. For $r/R \sim 0.1$ and $\phi_l = 0.2$, we expect $U_2 \sim 5 \text{ mm s}^{-1}$. Even though, $U_2 \gg U_1$, the droplet self-propulsion is dominated by the visco-capillary stage. This is because the droplet displacement s_2 during the second stage is negligible, since it is given by

$$\begin{aligned}
s_2 &= U_2 \tau_2 \\
&= \left(\frac{\phi_l}{\pi}\right)^{1/2} \left(\frac{r}{R}\right)^2 r
\end{aligned} \tag{S8}$$

, i.e., $s_2 \ll r, l$. Eq. (S7) also predicts $U_2 \propto 1/R^2$ a propulsion speed that decreases with increasing R , inconsistent with experimental results showing that U_L is independent of R .

Zoomed-in view of droplet self-propulsion

Using a microscope objective attached to a high resolution camera, we recorded the serpentine motion of droplets on lubricated surface ($\eta = 73 \text{ cP}$) over 4 hours of continuous cooling and characterized the self-propulsion speeds U_L for 40 different droplets with radii $R = 0.2\text{--}1.3 \text{ mm}$ [Fig. S6]. Figure S6(a)–(b) shows the serpentine motion for one such droplet with $R = 1.2 \text{ mm}$, with the neighbouring micron-sized droplets clearly resolved. In the green boxed region in Fig. S6(a), $\phi_l = 0.2$ and $\langle r \rangle = 18 \text{ }\mu\text{m}$.

We found that U_L is insensitive to R and $R/\langle r \rangle$, consistent with predictions in Eq. (S6). We also found that $\phi_l = 0.15\text{--}0.30$ (indicated by the blue band in Fig. S6(e)) remains constant throughout the 4 hours of experiment. Note that we measured ϕ_l in the vicinity but excluding the large droplet just before it swept through the region.

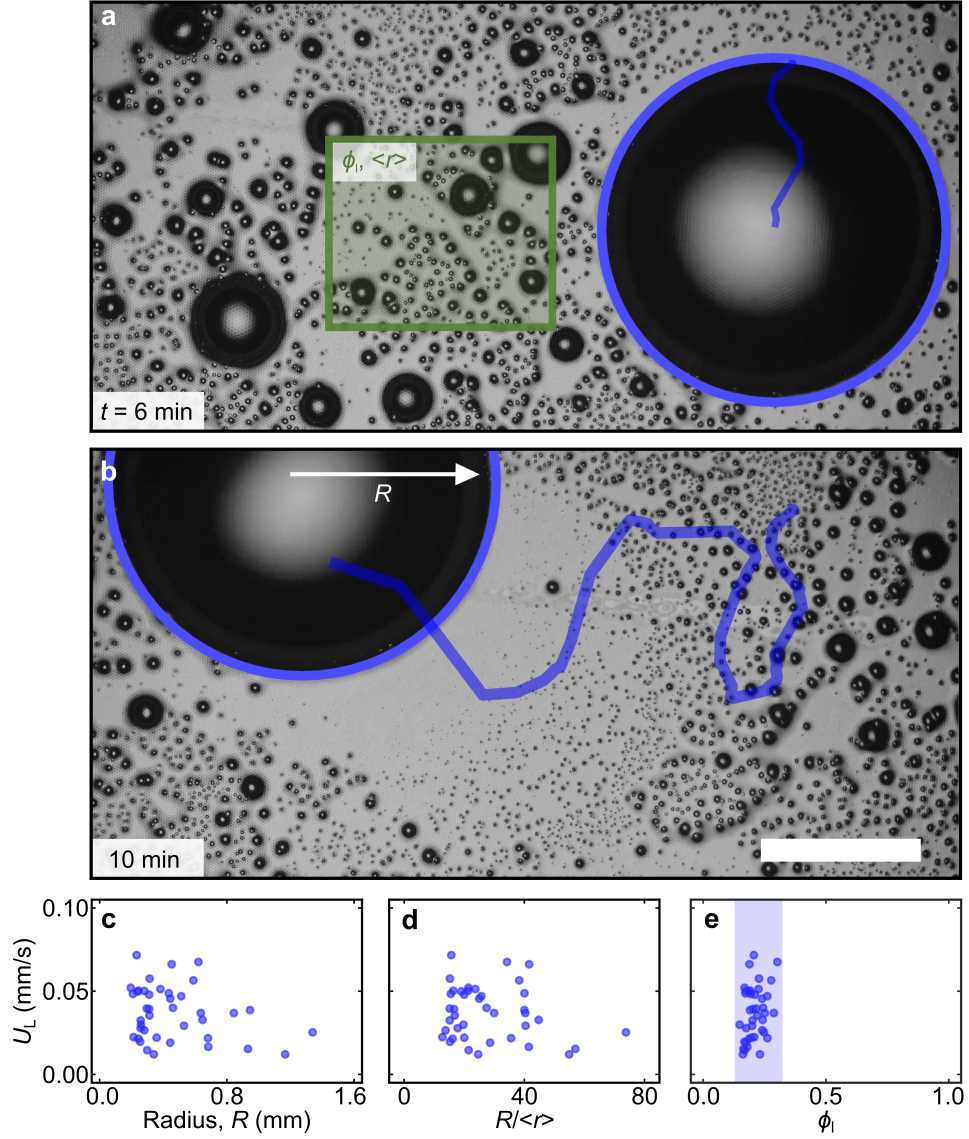


FIG. S6: (a)–(b) Serpentine motion a large droplet (circled blue) sweeping through much smaller droplets with mean radius $\langle r \rangle$ and surface coverage ϕ_l . Scale bar is 1 mm. We measured the self-propulsion speeds U_L for different (c) droplet radii R , (d) ratio of large and small droplets $R/\langle r \rangle$, and (e) local surface coverage ϕ_l over four hours of continuous cooling. Note that (c)–(e) are the same dataset as filled blue circles in Fig. 3(c) in the main text.

Surface coverage and droplet distribution

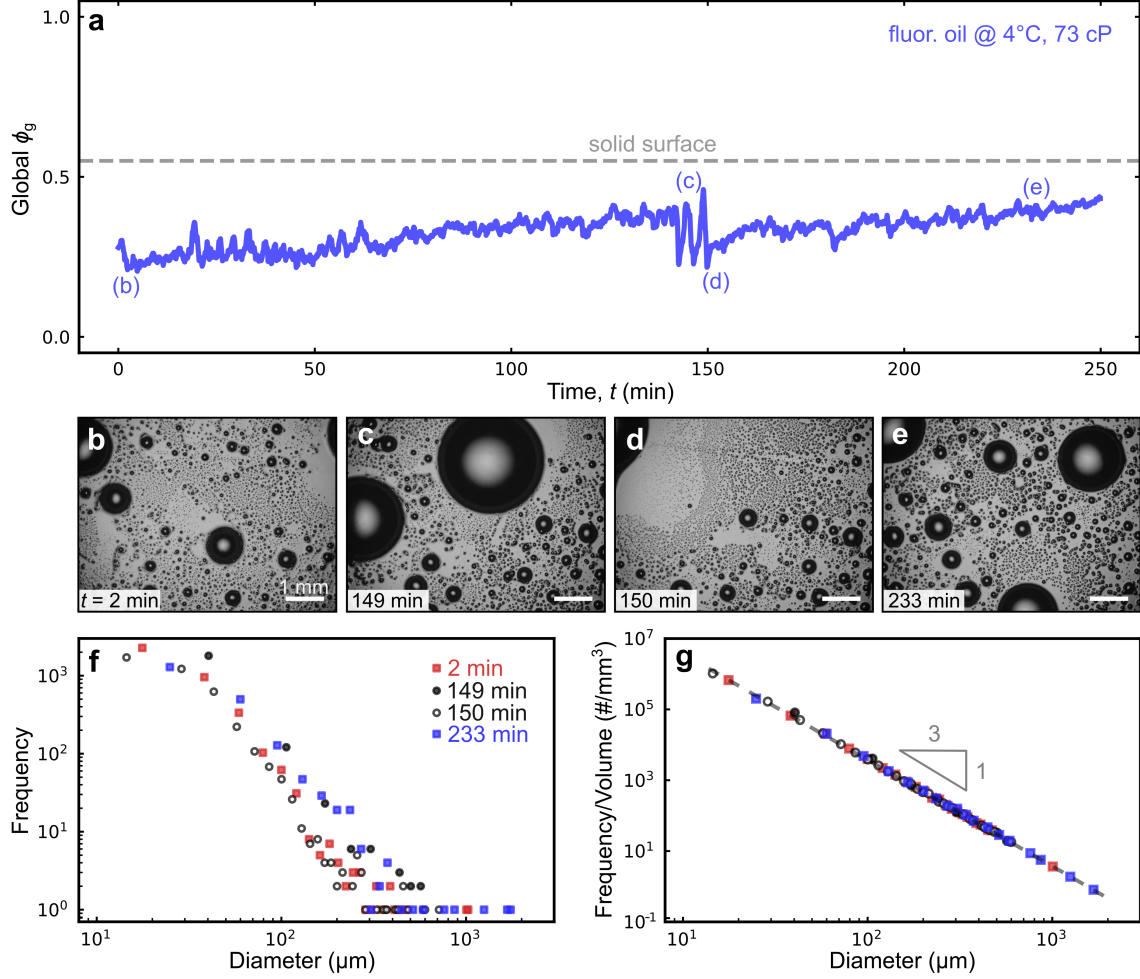


FIG. S7: (a) Global surface coverage ϕ_g as a function of cooling time. Note that the experiment excludes the first few minutes of cooling before ϕ_g equilibrates. (b)–(e) Snapshots at the timepoints $t = 2, 149, 150$, and 233 min. Scale bars are 1 mm. (f) Frequency distribution and (g) number density as a function of droplet diameter $2R$ at various timepoints correspond to (b)–(e). The dashed line shows the scaling of number density with diameter according to the power-law fit in Eq. (S9).⁵

In the previous section, the measured *local* ϕ_l excludes the large sweeping droplet. We have also measured the *global* surface coverage ϕ_g over the entire field of view of the camera ($6.7 \text{ mm} \times 5.1 \text{ mm}$) and found that $\phi_g = 0.33 \pm 0.05$ over the 4 hours of continuous cooling [Figs. 7(a)–(e)]. This saturation value of ϕ_g is slightly higher than that of ϕ_l and arises due to a complex interplay between the sweeping motion of the large droplets and

the recondensation rates. Interestingly, ϕ_g for lubricated surfaces is significantly lower than the value of 0.55 typically encountered on solid surfaces.⁶ Note that large fluctuations of ϕ_g at around $t = 150$ min are due to sweeping motion of large droplets as evident from the snapshots in Figs. 7(c), (d). We also found that the number density function $N(R)$ (frequency/volume) for droplets on lubricated surfaces follow the power law distribution

$$N(R) = aR^{-b} \tag{S9}$$

with constants a and b are fitting parameters. Fitting the experimental results with Eq. (S9) [Fig. S7(g)], we obtained $b = -3$ which is consistent with the results reported by Weisensee et al. (2017).⁵ As time passes, the polydispersity of the droplet sizes increases, as discussed in Fig. 1(b) in the main text [Figs. 7(f), 7(g)].

Hot vapour condensates

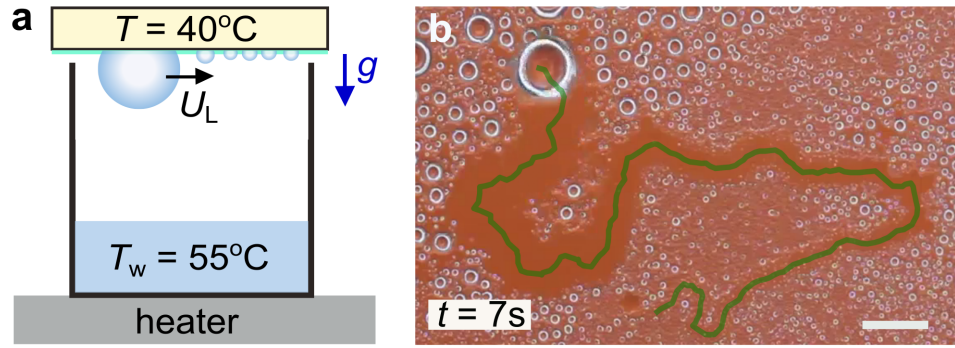


FIG. S8: *Self-avoiding motion of hot vapour condensate.* (a) Schematic and (b) photograph (0.5 mm scale bar) showing similar self-avoiding droplet motion from hot vapour condensing on lubricated nanostructured surface.

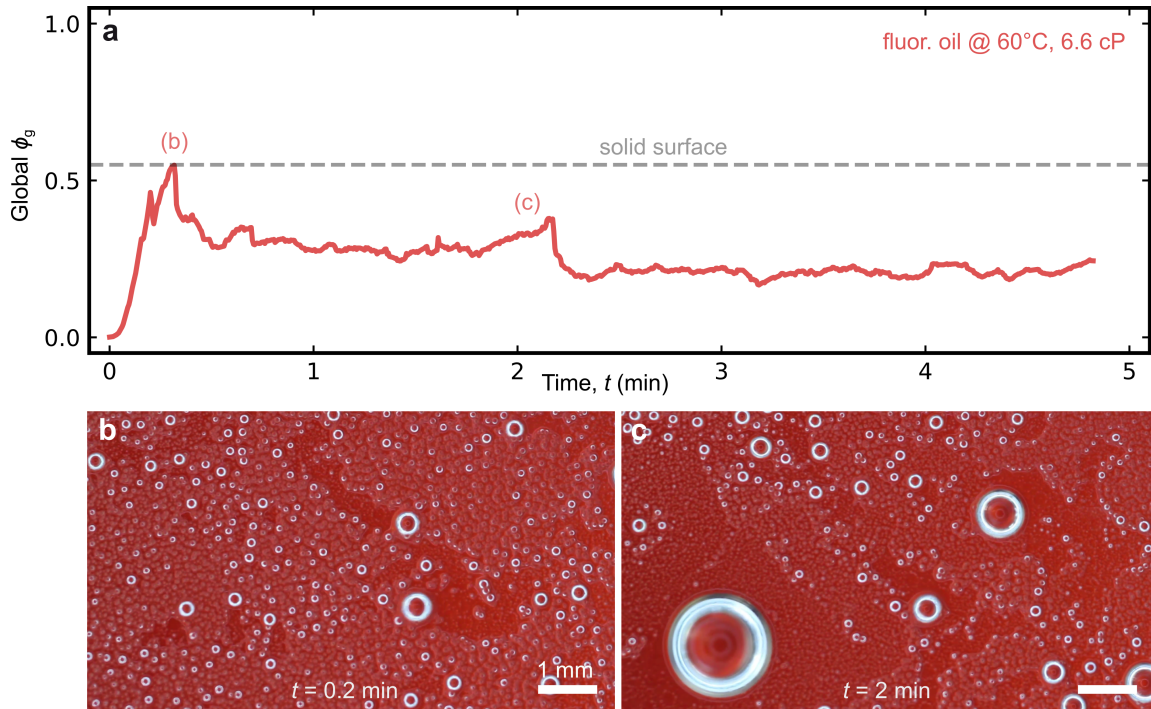


FIG. S9: (a) Global surface coverage ϕ_g as a function of time for hot vapour condensates. (b)–(c) Snapshots at timepoints $t = 0.2$ and 2 min.

For hot vapour condensates, ϕ_g similarly saturates at 0.24 ± 0.05 , below the typical 0.55 value typically encountered on solid surfaces [Fig. S9].

Circular motion

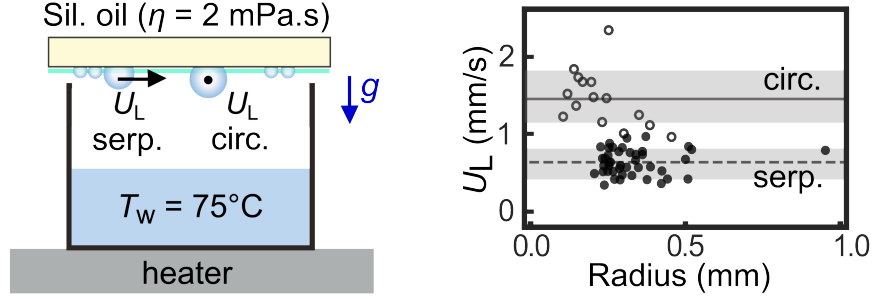


FIG. S10: U_L as a function of droplet radius for serpentine and circular motions. Dashed and full lines indicate the mean velocities, while gray regions represent the standard deviations.

As discussed in the main text, in the lubricant-depleted regions, there are small pockets with thicker lubricant overlayer where droplets ($R = 0.1\text{--}0.5 \text{ mm}$) remain mobile and move in circular motions. Interestingly, we found that the exact magnitude of U_L depends on the motion types. In Fig. S10, $U_L = 1.4 \pm 0.4 \text{ mm s}^{-1}$ for circular motion is higher than the corresponding $U_L = 0.6 \pm 0.2 \text{ mm s}^{-1}$ for serpentine motion. More importantly for both motions, U_L is relatively insensitive to droplet radius R consistent with Eq. (S6).

Lubricant-depleted regions

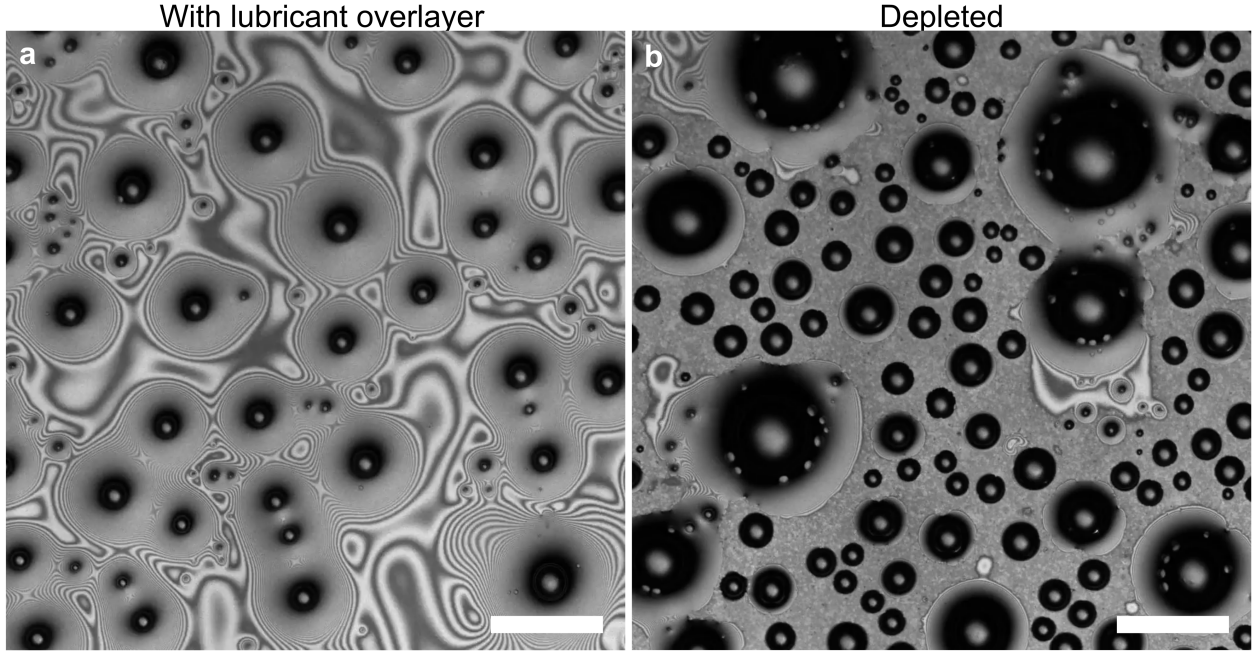


FIG. S11: Menisci around individual droplets when lubricant film is (a) pristine and (b) depleted, observed using reflection interference contrast microscopy as described in Fig. S1. Wherever there is a lubricant overlayer, we observe bright and dark fringes corresponding to different lubricant heights. Scale bars are 1 mm.

We used reflection interference contrast microscopy to confirm the presence or absence of lubricant overlayer. Wherever there is a lubricant overlayer, we observe bright and dark fringes [Fig. S11(a)]. Wetting ridges form around the droplets, and importantly these ridges can overlap and create capillary attraction between neighbouring droplets, driving the collective motion described in the main text.

As time passes, the motion of the sweeping droplets locally depletes the lubricant film. As discussed in our previous publication,⁷ this is driven by the wetting ridge of the sweeping droplets. In Fig. S11(b), only the largest droplets retain enough lubricant to create wetting ridges. Importantly, the wetting ridges cannot overlap effectively and therefore the droplets remain stationary.

Once the lubricant is depleted, there is no longer serpentine motion or interdroplet attraction [Supporting Video S9; cf., Supporting Video S5]. For the cooled experiment in Fig. 3, this occurs after more than 4 hours of experiment ($t = 250$ min). At which point, the global ϕ_g starts climbing to the 0.55 value normally encountered on solid surface [Fig. S12].

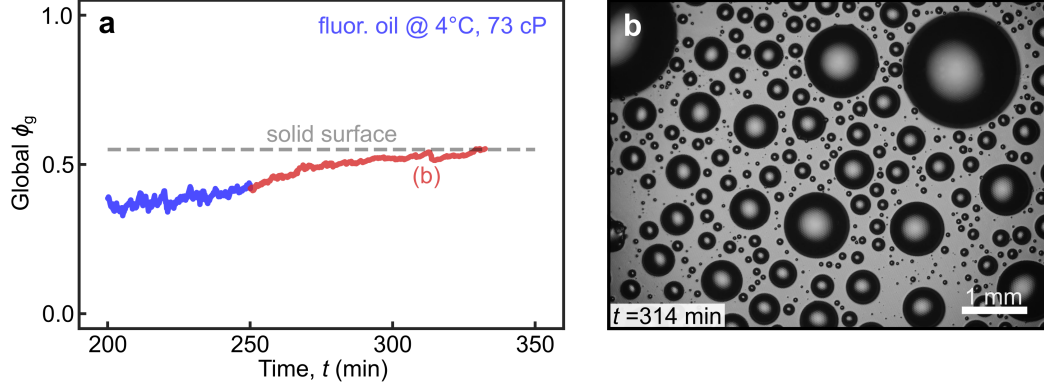


FIG. S12: (a) Global surface coverage ϕ_g as a function of cooling time. Lubricant depletion occurs at $t = 250$ min. (b) Snapshot at timepoint $t = 314$ min, where $\phi_g = 0.52$. Scale bar is 1 mm.

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