Supporting Information

Color and Texture Morphing with Colloids on Multilayered Surfaces

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Contents  
- Figure. S1-9  
- Note 1  
- References
Figure S1. Illustration of morphing over a large area for complicated pattern. Randomly dispersed glass beads in the liquid top layer (A) can selectively cover many places of green patches to show a cross-like pattern (B, C) after exposure to red laser light.
Figure S2. Solar cell panel as the solid substrate. (A) Photograph of two commercial amorphous silicon (a-Si) panels with (rear) and without (front) the aluminum top electrode; (B) Configuration of the a-Si solar panel; (C) Optical microscope image of three adjacent channels on the solar panel without the top Al film; and (D) AFM image of the a-Si (thickness of 700 nm).
Figure S3. Absorption and reflection of an incident laser (405 nm) by the a-Si substrate. The a-Si film absorbs 76.2% of light. If we assume a Gaussian distribution of the light absorption profile ($e^{-cx}$), when $x = 700$ nm the calculated absorption coefficient ($c$) is $2.05 \times 10^{-3}$ nm$^{-1}$. 
Figure S4. Very low intensity light can trigger liquid eruption. 2-BE (125µm)/solar panel was exposed to 405 nm laser beam of 0.02 W cm\(^{-2}\) power intensity. A concave mirror in 2-BE film formed after an exposure of 25 minutes, where two reflected laser spots changed from small dots into large ellipses. Textures inside the rings are Newton rings when the thickness of the film became comparable to the wavelength of incident light.
Figure S5. Cycled responses of our multilayered thin films to pulsed lasers. The sequences were recorded by a photodiode and an oscilloscope.
Figure S6. Thickness of TOPO. Optical microscope image of the solid TOPO film in Figure 4. One piece of TOPO was placed between two glass slides for thickness measurement.
Figure S7. Light-triggered texture morphing in an all-solid system setup. Hot plate was used to heat the TOPO covered thin films. Laser pointer and photomask were used to generate patterns. The motion of laser was controlled by a translation stage on the right.
Note S1. Motion of particles under the influence of a thermal gradient.

1.1 The relationship between particle speed ($v_r$) and the thickness ($h$) of the liquid layer

Surface tension along liquid/air interface is sensitive to temperatures, in which a spatial variation in temperature can induce nonuniform tensions. This uneven distribution over a liquid surface can further trigger tangential stress, resulting in fluid motion dubbed as thermocapillary convection or Maragoni flow. In our experiment (see Figure 2, Figure S8), a thermocapillary convection is induced by a laser illumination: absorbed laser heats up the substrate and the heat transfers to the liquid above; and therefore a surface tension gradient is generated along the liquid/air interface. If we assume the total heat supplied by the bottom substrate is consumed by the whole liquid above, the generated thermal gradient $\sigma$ is then inversely proportional to the thickness of the liquid layer:

$$\sigma \propto \frac{q}{h} \quad \text{(s1)}$$

where $q$ is the laser intensity. On the other hand, we know that the surface tension is inversely proportional to the temperature. Hence, the surface tension gradient is proportional to the thermal gradient but with an opposite direction (Figure S8). If we only consider the magnitude of the surface tension gradient ($\tau$), we have:

$$\tau \propto \frac{q}{h} \quad \text{(s2)}$$

For an incompressible and isotropic Newtonian fluid, like 2-BE in our experiment, the surface shear stress (the surface tension gradient in our case) is proportional to the velocity gradient along the direction perpendicular to the surface. By assuming a velocity profile shown in Figure S8, where the thickness ratio between the left-ward flow and the right-ward flow, $\alpha$, is a constant and not dependent on the liquid thickness, we have:

$$\tau \propto \frac{v_2}{a h} \quad \text{(s3)}$$

Substituting s3 back into s2, one can obtain that:

$$v_2 \propto \alpha q \quad \text{(s4)}$$

which shows that the liquid flow speed at the top surface is independent of the thickness of the liquid layer.

The bottom fluid follows the direction of the thermal gradient, which drives particle move towards the center of the high temperature area, and eventually manipulates the particles to form one monolayer. To conserve all the masses, the areas in the blue and yellow zones need to match:

$$(1 - \alpha) h \cdot v_1 = \alpha h \cdot v_2 \quad \text{(s5)}$$

From the velocity profile shown in the Fig. S7, we can also obtain that:

$$\frac{v_1}{\beta h} = \frac{v_2}{a h} \quad \text{(s6)}$$

$$\frac{v_r}{r} = \frac{v_1}{(1 - \alpha - \beta) h} \quad \text{(s7)}$$

Combining Equation (s4-s7), we have:

$$v_r \propto \frac{r q}{h} \quad \text{(s8)}$$

which in the end tells us that the particle velocity is inversely proportional to the layer thickness, but proportional to the laser intensity.
Figure S8. Velocity profile of the liquid flow induced by the laser. The upper flow is with an opposite direction of the thermal gradient, while the bottom flow, pushing the particles towards the illumination center, is of the same direction as the thermal gradient.

1.2 The time ($t$) to produce a well inside the liquid
During the Stage 2 in Figure 2B, we illustrated that a liquid well will form after a brief laser exposure. This process is made possible by the surface shear force along the liquid/air interface. Assume the work done by this shear force all converts into the gravity potential of the liquid, and assume the liquid well is of a cylinder in shape. We have the total gravity potential:

$$P = \rho \cdot h \pi R^2 \cdot g \cdot \frac{h^2}{2}$$  \hspace{1cm} (s9)

where $R$ is the radius of the well (roughly equals to the size of the laser spot), $\rho$ as the liquid density, $g$ for gravity acceleration. The work done by the shear force (surface tension gradient):

$$W = \tau \cdot \pi R^2 \cdot v_2 \cdot t \propto \frac{q^2}{h} \cdot \pi R^2 \cdot v_2 \cdot t$$

Since $W = P$, and $v_2$ is linearly dependent on the laser intensity ($q$), the following relationship can be derived:

$$t \propto h^3 \text{ and } t \propto q^{-2}$$

Figure S9 showed our experimental data points that fit very well with the above relationships.

Figure S9. The inverse second order relationship between $t$ and $q$ is verified by experiment results (red line and points). The thickness of the liquid layer is around 125 $\mu$m. Both
experimental results and the modeling show that $t$ is proportional to the cube of the liquid thickness. The laser intensity $q$ is fixed at 0.4 W·cm$^{-2}$ to determine the thickness dependence.

Similar as solid thin film dewets when its thickness reaches a small value,$^2$ for the discussed liquid rupture under a light exposure (Figure 2A), a critical thickness ($d_c$) also exists. This $d_c$ depends on the complex interplay between two energies, they are, liquid/air interface energy and liquid/substrate energy. When laser is used, parameter $d_c$ also depends on the power intensity of the incoming light. In our experiments, we observed a $d_c$ for our liquids to be around 200 µm when the light intensity is 0.02 W·cm$^{-2}$. Beyond this $d_c$ value, no obvious liquid rupture or turbulence is observed.

References