Electronic Supplementary Material (ESI) for Soft Matter

Supplementary Information for

Breathing, crawling, budding, and splitting of a liquid droplet under laser heating

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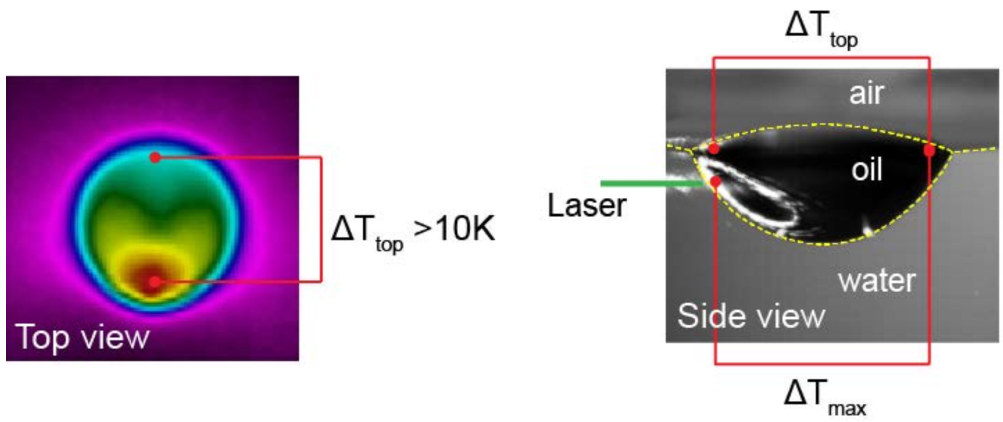
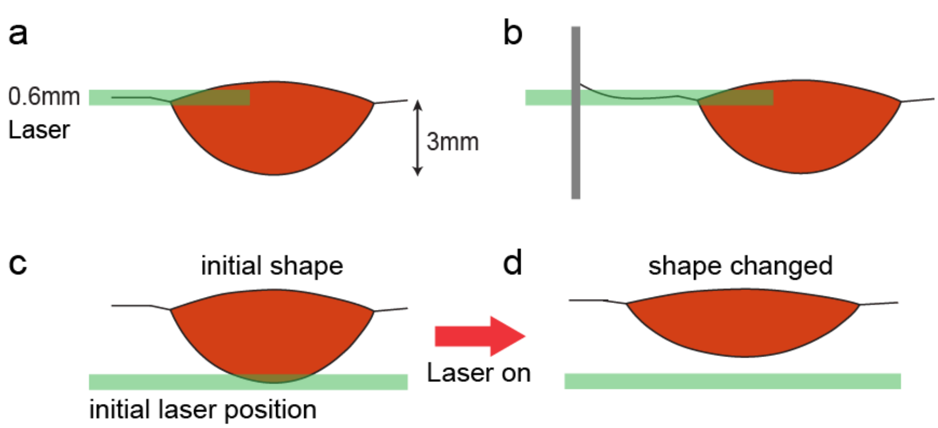
	Nitrobenzene oil	Aqueous solution
Thermal conductivity, k (W/m K)	**0.163**	0.596
Specific heat capacity, Cp (J/kg K)	**1400**	4181
Kinetic viscosity, ν (m^2/s)	**1.4 E-06**	0.89 E-06
Coefficient of thermal expansion, α (1/K)	**0.8 E-03**	0.2 E-03
Density, ρ (kg/m^3)	**1230**	1000
Rayleigh Number, *Ra*, at ΔT=4K and h=2mm	$Ra \equiv \frac{\alpha g \Delta T h^3}{D_T \nu} = \frac{\alpha g \Delta T h^3 \rho C_p}{k \nu} = 1811$	

Supplementary Table 1 | Rayleigh Number *Ra*. In Rayleigh-Bénard convection, the initial movement to form convection cells is the upwelling of lesser density liquid from the heated bottom layer. The instability sets in, and convection flow appears above a critical Rayleigh number Ra_c. The Ra_c for two rigid and free boundary conditions are known 1708 and 657, respectively. When ΔT = 4K and h = 2mm, *Ra* of nitrobenzene droplet is larger than the Ra_c values for both boundary conditions. In this experiment, ΔT that measured from the temperature distribution at the top surface of droplet using a thermal camera was larger than 10K. (See Supplementary Figure 1)



Supplementary Figure 1 | Temperature differences. ΔT_{top} is a temperature difference at the top surface of the droplet, and ΔT_{max} is the ideal maximum temperature difference inside droplet. In our system, ΔT_{top} is larger than 10K.



Supplementary Figure 2 | Laser beam size effect. It is quite difficult to define h/H for the cases of the top and bottom heating. In the case of the top heating (h/H~0), due to the finite size of the laser beam, only part of the laser beam is shined on the droplet surface (see (a)). And some part of the laser beam becomes scattered a lot around container/water/air contact line (see (b)). These make it difficult to define h/H Fig. 7(b). In the case of bottom heating (h/H~1), it also has the beam size effect as at the top heating case (see (c)). And also the shape change of the droplet makes it difficult to define h/H (see (d)).

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Supplementary Movie 1 | Motion and shape in the breathing mode. Top and side views of a floating droplet at a low laser power below 0.3W. The droplet is periodically deformed while in fixed position. The bottom of the droplet moves periodically up and down, as the diameter of the droplet oscillates simultaneously to conserve the total droplet volume.

Supplementary Movie 2 | Convection flow in the breathing mode. Top and side views of a floating droplet at a low power laser below 0.3W. In order to observe the flow inside the droplet, a small number of hydrophobic colloidal tracer particles are added to the droplet.

Supplementary Movie 3 | Temperature distribution in the breathing mode. Temperature distribution at the top surface of the droplet at a low power laser below 0.3W. According to this movie the temperature distribution changes periodically; the convection occurs momentarily and ceases during the rest of the time within each period, and there exists a small high-temperature spot for a short moment within each period in the steady condition.

Supplementary Movie 4 | Motion and shape in the crawling mode. Top and side views of a floating droplet. At a low power laser around 0.3W, the oscillating droplet begins to move toward the laser. As the laser power is increased over 0.33W, the droplet moves toward the laser without the oscillation.

Supplementary Movie 5 | Convection flow in the crawling mode. Top and side views of a floating droplet at a low power laser near 0.3W. The convection flow is continuous in time while the droplet moves with constant speed in the steady condition.

Supplementary Movie 6 | Temperature distribution in the crawling mode. Temperature distribution at the top surface of the droplet at the laser power near 0.3W. After the initial transient period, the surface temperature distribution is uniform, suggesting the establishment of the steady convective flow inside the droplet.

Supplementary Movie 7 | Motion and shape in the budding mode. Top and side views of a floating droplet at the laser power near 1.5W. The droplet crawls with periodic budding, in which a sharp protrusion appears for a short time at the heated region and soon retracts**.** As the laser power is increased further, the protrusion extends longer and begins to form a satellite droplet after several buddings. But the still-connected satellite droplet retracts eventually and recombines with the mother droplet.

Supplementary Movie 8 | Motion and shape of in the splitting mode. Top and side views of a floating droplet at the laser power higher than that of the budding mode. When the satellite droplet grows sufficiently large, it detaches from the mother droplet. During the splitting mode, the mother droplet is pushed back from its initial position to conserve momentum.