Light Induced Surface Tension Gradients for Hierarchical Assembly of Particles from Liquid Metals

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Achieving control over the motion of dissolved particles in liquid metals is of importance for the meticulous realization of hierarchical particle assemblies in a variety of nanofabrication processes. Brownian forces can impede the motion of such particles, impacting the degree of perfection that can be realized in assembled structures. Here we show that light induced Marangoni flow in liquid metals (i.e., liquid-gallium) with Laguerre-gaussian (LG_{pl}) lasers as heating sources, is an effective approach to overcome Brownian forces on particles, giving rise to predictable assemblies with high degree of order. We show that by carefully engineering surface tension gradients in liquid-gallium using non-gaussian LG_{pl} lasers, the Marangoni and convective flow that develops in the fluid drives the trajectory of randomly dispersed particles to assemble into 100-μm wide ring-shaped particle assemblies. Careful control over the parameters of the LG_{pl} laser (i.e., laser mode, spot size, and intensity of the electric field) can tune the temperature and fluid dynamics of the liquid-gallium as well as the balance of forces on the particle. This in turn can tune the structure of the ring-shaped particle assembly with a high degree of fidelity. The use of light to control the motion of particles in liquid metals represents a tunable and rapidly reconfigurable approach to spatially design surface tension gradients in fluids for more complex assembly of particles and small-scale solutes. This work can be extended to a variety of liquid-metals, complementary to what has been realized in particle assembly out of ferrofluids using magnetic fields.

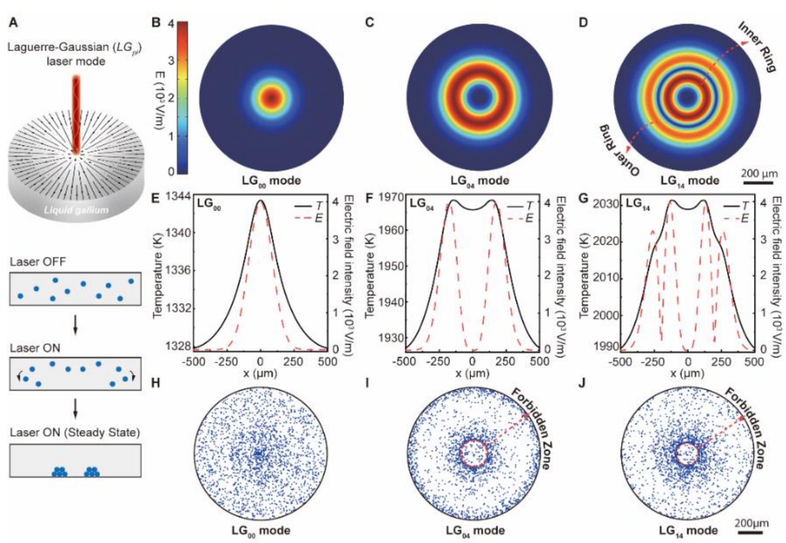

Figure 1 (A) Schematic highlighting the process of assembling particles out of liquid gallium using a LG_{pl} laser mode. (B, C and D) Profile of the electric field on the surface of liquid gallium under (B) LG_{00}, (C) LG_{04}, and (D) LG_{14} laser modes, respectively. (E, F and G) Profiles of the electric field (*dashed red lines*) and temperature (*solid black lines*) on the surface of liquid gallium along the x-axis. (H, I and J) Resulting particle patterns at the liquid-solid interface for (H) LG_{00}, (I) LG_{04}, and (J) LG_{14} laser modes interacting with liquid gallium, respectively.

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Suplementary Pages

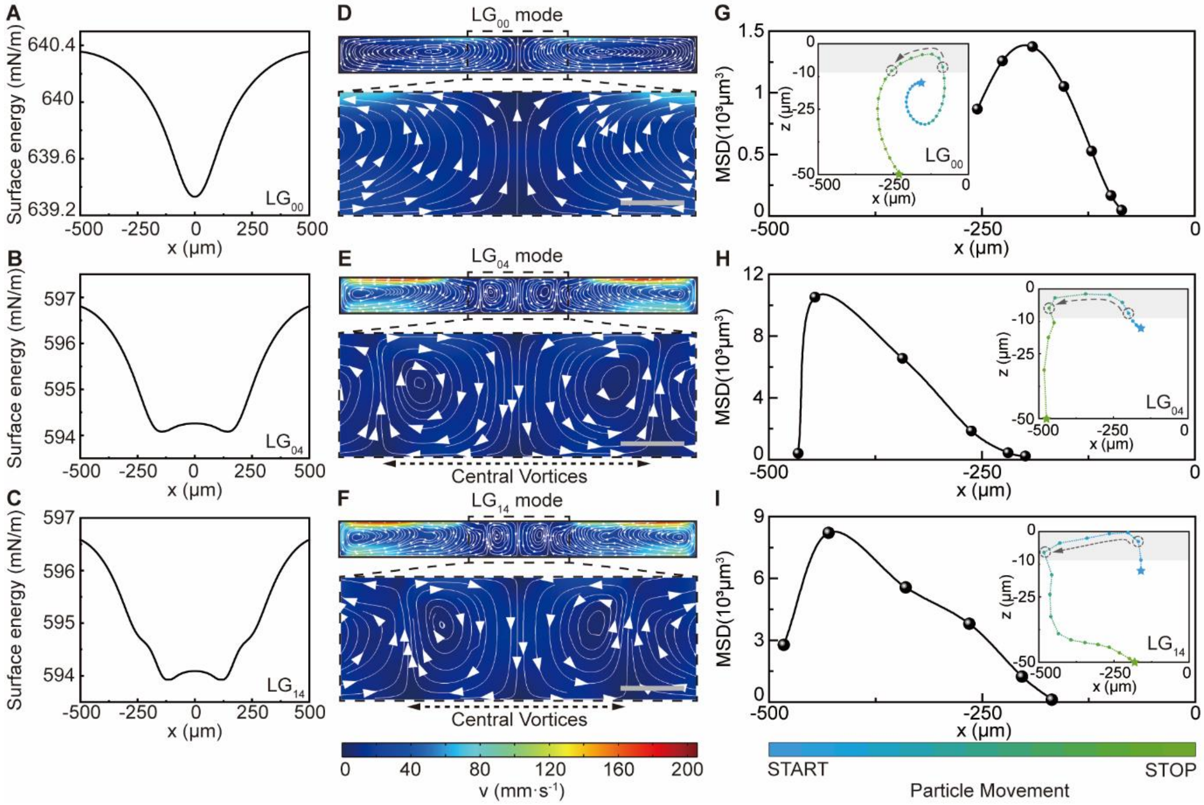

Figure 2. Influence of surface tension gradients of liquid gallium on the particle motion in the bulk fluid. (A, B, and **C)** Surface tension profiles of liquid gallium along the x-axis (i.e., y-axis and z-axis = 0) for **(A)** LG_{00}, **(B)** LG_{04}, and **(C)** LG_{14} laser modes ($\lambda = 645\ nm$ and $w_o = 125\ \mu m$), respectively. **(D, E,** and **F)** Convective flow patterns induced by the surface tension gradient (i.e., Marangoni flow) for **(D)** LG_{00}, **(E)** LG_{04}, and **(F)** LG_{14} laser modes, respectively. For **(E)** LG_{04}, and **(F)** LG_{14} laser modes, two additional central vortices are observed in the convective flow patterns in the fluid, as highlighted by *dashed black lines* under the zoomed in regions. The scale bar for all zoomed in regions is 50 μm. The absolute value of fluid velocity is indicated by the color bar at the bottom. **(G, H,** and **I)** The mean-squared displacement (MSD) for particles under **(G)** LG_{00}, **(H)** LG_{04}, and **(I)** LG_{14} laser modes when particle moves near the top surface of liquid gallium ($|z| \leq 10\ \mu m$, *shaded gray background*). The insets in **(G, H,** and **I)** are the particle trajectories (*blue-to-green gradient*) guided by the fluid flow induced under the **(G)** LG_{00}, **(H)** LG_{04}, and **(I)** LG_{14} laser modes, respectively. All particles were released at the same position in the fluid (*blue star*) and finally assembled at liquid-solid interface (*green star*). The data points used for the MSD analysis are highlighted with *dashed black circles* and *dashed black arrows*.

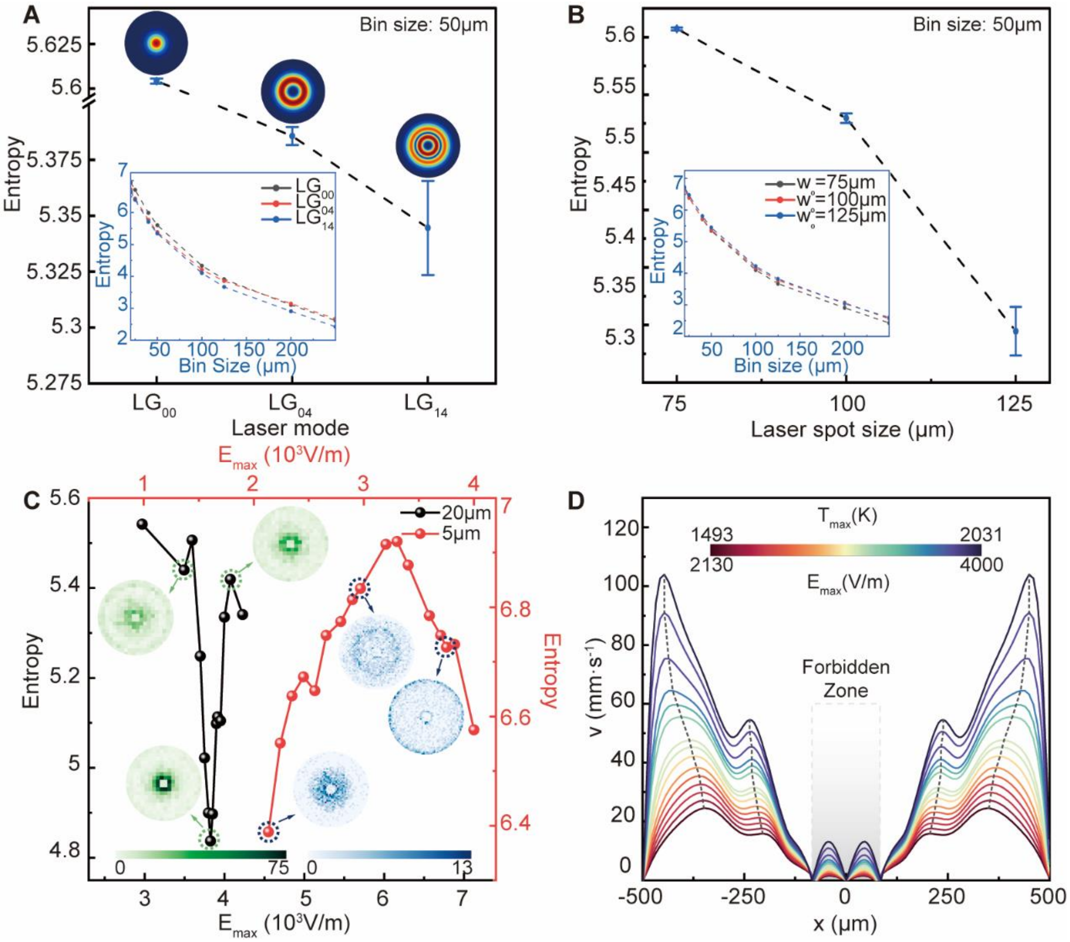

Figure 3. Summary of the tuning knobs of LG_{pl} laser modes to obtain ring-shaped pattern with different degrees of order. (A) Impact of LG_{pl} laser modes: entropy of particle patterns ($d_p = 20\ \mu m$) under different LG_{pl} laser modes (LG_{00}, LG_{04}, and LG_{14} laser modes, $w_0 = 125\ \mu m$, and $E_{max} = 4000\ V/m$). Bin size for the calculation of entropy is $50\ \mu m$. To avoid the influence of bin size, the entropy with different bin sizes is shown in the inset. **(B)** Impact of LG_{pl} laser spot size: entropy of particle patterns ($d_p = 20\ \mu m$) under LG_{14} laser mode ($E_{max} = 4000\ V/m$) with spot size of $75\ \mu m$, $100\ \mu m$, and $125\ \mu m$. Bin size for the calculation of entropy is $50\ \mu m$. The entropy with different bin sizes is also shown in the inset. **(C)** Impact of maximum intensity value of the electric field: entropy of particle patterns under different E_{max} values (LG_{14} laser mode and $w_0 = 125\ \mu m$). The bin size for the calculation of entropy is $50\ \mu m$ for $20\ \mu m$ diameter particles (*solid black line*), and the bin size is $20\ \mu m$ for $5\ \mu m$ diameter particles (*solid red line*). Corresponding density distribution maps for selected data points (*dashed circles* in **(C)**) are shown. The count of particles in each square grid is indicated by the color bar at the bottom (*green-to-white gradient* for $20\ \mu m$ diameter particles, and *blue-to-white gradient* for $5\ \mu m$ diameter particles). **(D)** Fluid velocity profile along solid-liquid interface under E_{max} values. The velocity peaks are marked by *dashed black line*. The area of "forbidden zone" is highlighted in *shaded grey* background. E_{max} and corresponding temperature maxima (T_{max}) in the liquid gallium are indicated by the top color bar.