Supporting Information

Directional Leidenfrost Droplet Propulsion on Femtosecond-Laser-Engineered Dual Heterogeneous Surfaces with Hybrid Boiling States

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Figure S1-S7

Captions of Supplementary Movies

Movie S1. Wettability and spreading process of a water droplet on the untreated smooth aluminum substrate and the laser-structured aluminum surface.

Movie S2. Unidirectional transport of water droplet on the heated aluminum sheet with two different laser-structured regions.

Movie S3. Influence of the heating temperature on the dynamic behavior of the deposited droplets on the structured aluminum surface.

Movie S4. Unidirectional droplet propulsion of various volatile liquids on the heated structured aluminum surface.

Movie S5. Details showing the generation of numerous tiny bubbles inside the droplet in the process of droplet propulsion.

Movie S6. Details showing the ejection of numerous tiny satellite droplets in the process of droplet propulsion.

Movie S7. Comparison of droplet dynamics on the unablated smooth aluminum surface and the fully laser-structured surface at a heating temperature of 250℃.

Movie S8. Multifunctional droplet propulsion and applications (transport along a curved path, droplet expulsion, droplet trapping, and droplet rotor).

Supplementary Figures

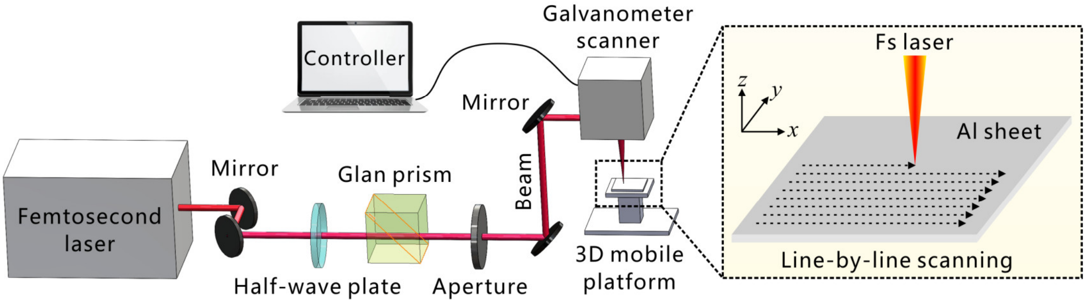

Figure S1. Diagram of the femtosecond laser processing system.

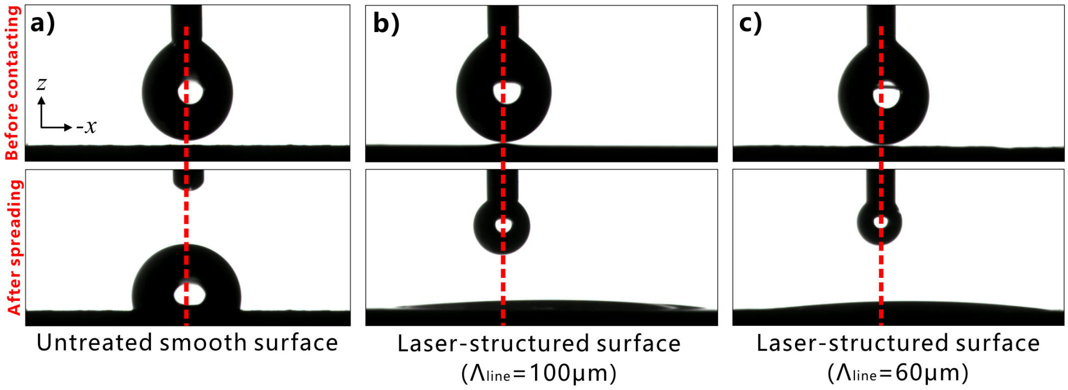

Figure S2. Wettability and spreading process of a water droplet on the (a) untreated smooth and (b,c) laser-structured aluminum surface.

When a water droplet comes into contact with a smooth aluminum surface, it adheres to the material surface, forming a symmetrical hemispherical shape (Figure S2a). In contrast, when the droplet contacts the laser-structured surface, it rapidly spreads across the material surface, ultimately exhibiting superhydrophilicity with a contact angle of less than 10° (Figure S2b and S2c). Notably, when using the syringe needle axis as the central axis, the droplet spreading demonstrates asymmetry. The spreading distance along the -*x* direction is greater than that along the +*x* direction, indicating that the droplet preferentially spreads toward the -*x* direction.

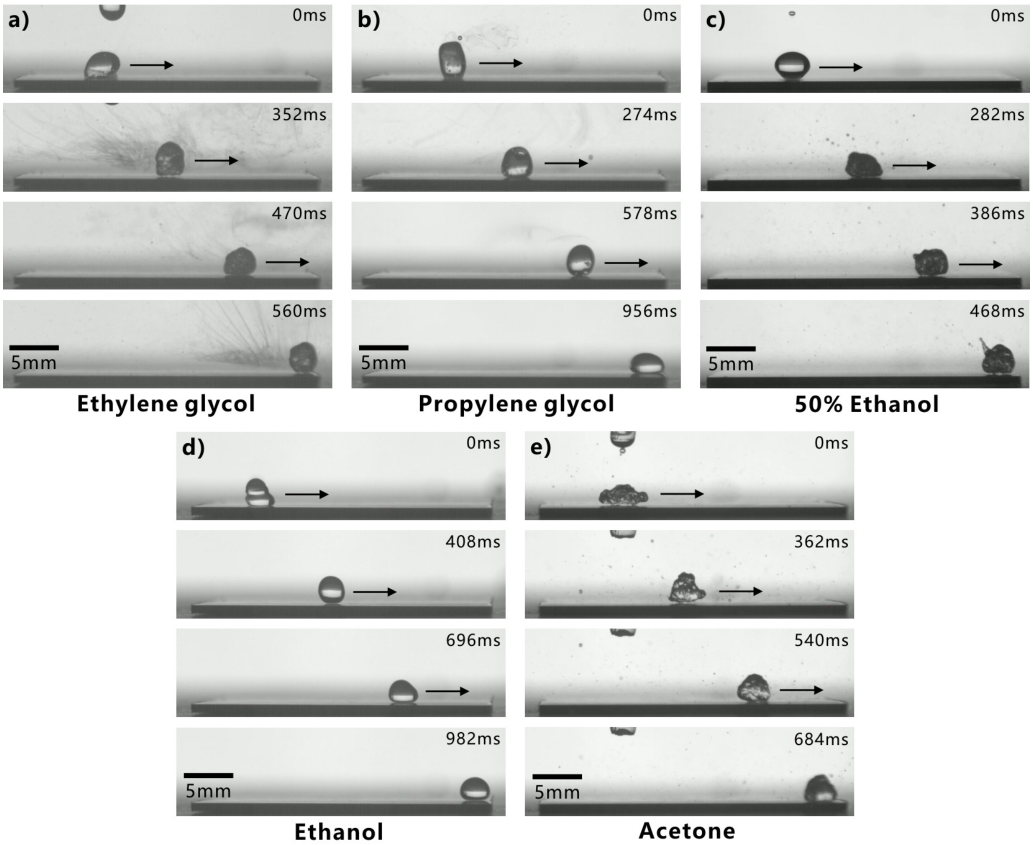

Figure S3. Process of unidirectional droplet propulsion of various volatile liquids on the heated structured aluminum surface: (a) ethylene glycol droplet at 310℃, (b) propylene glycol droplet at 320℃, (c) 50% ethanol droplet at 200℃, (d) anhydrous ethanol droplet at 305℃, and (e) acetone droplet at 110℃.

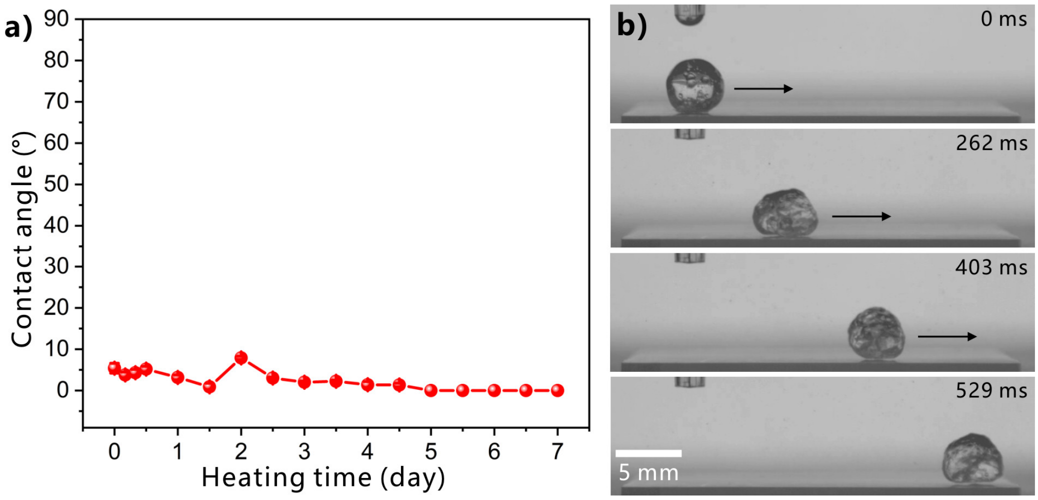

Figure S4. Effect of heating duration (at 260°C) on the wettability of laser-structured surfaces and the characteristics of directional proplusion of Leidenfrost droplets. (a) Variation in contact angle of water droplets (at room temperature) on the sample surface with increasing heating time. (b) Directional proplusion process of water droplets on the heated surface after seven days of heating treatment.

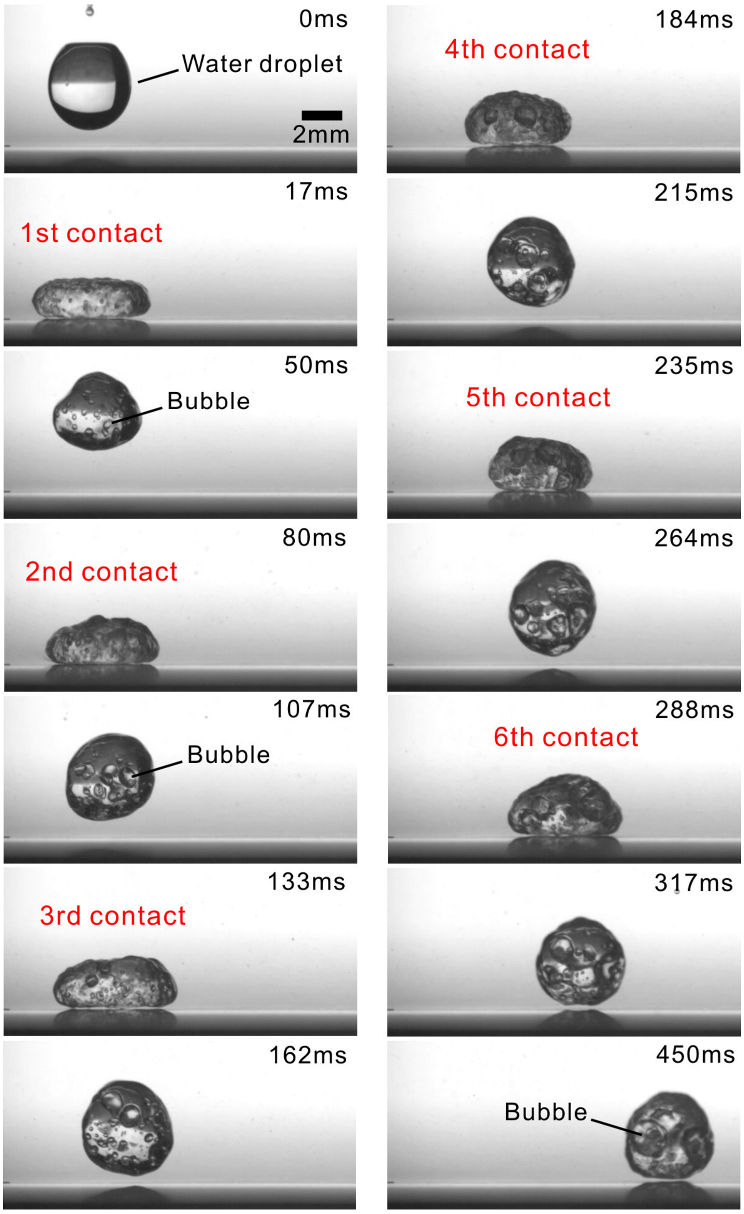

Figure S5. Details of the initial contact–rebound process of a water droplet falling onto a heated structured surface. From these images, it can be observed that when the droplet comes into contact with the hot surface, numerous tiny bubbles are generated inside the droplet. Over time, the number of bubbles decreases, but their size increases.

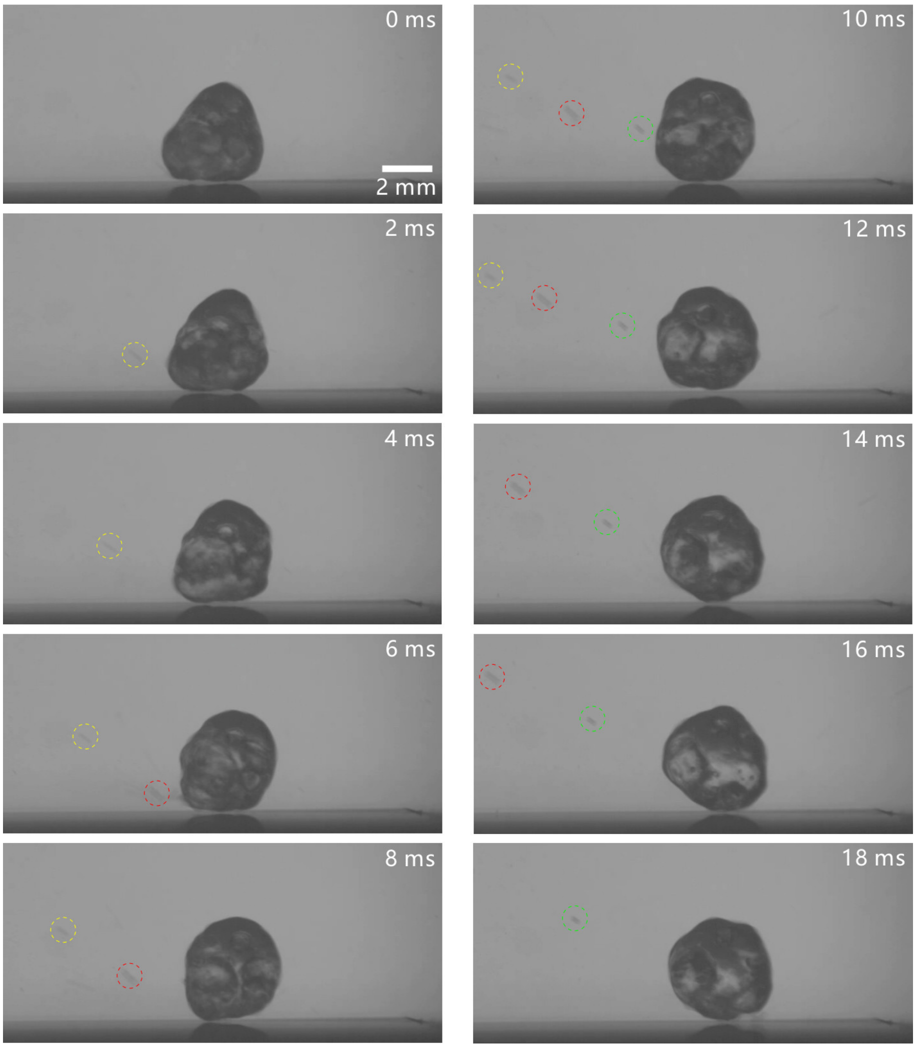

Figure S6. Sequence of snapshots captured by a high-speed camera showing the ejection of numerous tiny satellite droplets in the process of a water droplet moving on the heated sample surface. The time interval between consecutive snapshots is 2 ms. The spatial position changes of three ejected small satellite droplets are illustrated by the yellow, red, and green dashed circles.

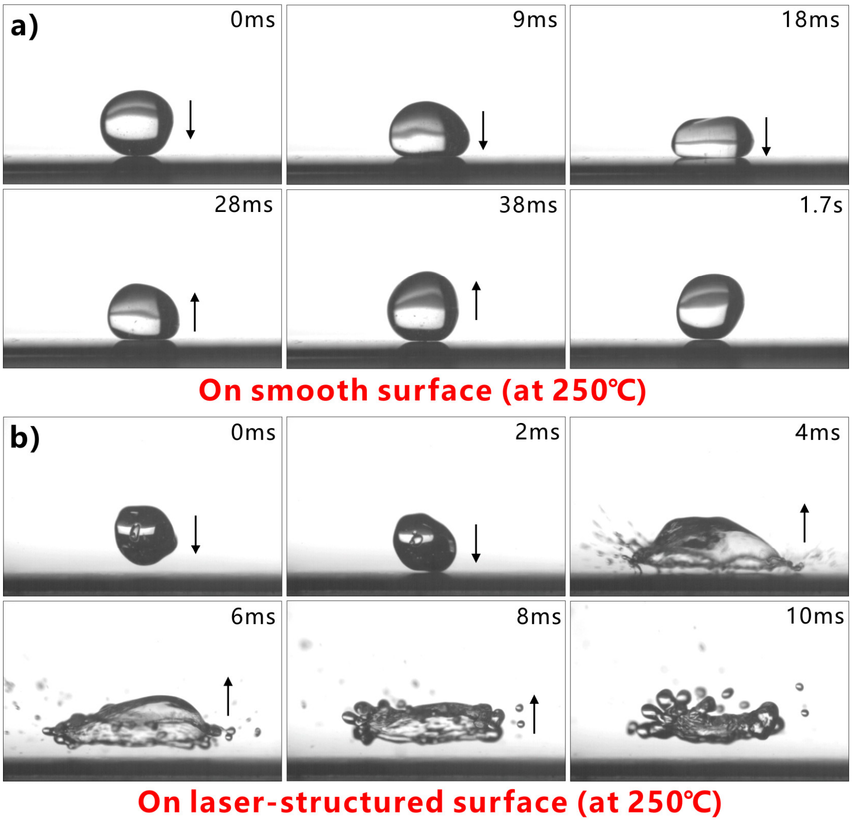

Figure S7. Comparison of droplets on the unablated substrate and the laser-structured microstructures at a heating temperature of 250°C: (a) on original smooth aluminum surface without laser-induced microstructures and (b) on fully laser-structured surface (Λ_{crater} = 30 μm, Λ_{line} = 10 μm).

When the heating temperature is set at 250°C, droplets deposited on untreated smooth aluminum surfaces exhibit the Leidenfrost state (Figure S7a). Although the droplets undergo spontaneous vertical oscillations, they maintain a stable spherical shape on the surface. After the self-induced oscillations subside, a stable translucent layer becomes visible beneath the droplet on the substrate, attributed to the vapor layer formed between the droplet and the heated substrate. In contrast, when droplets are deposited on surfaces fully covered with laser-induced microstructures, an intense phase transition occurs immediately upon contact, accompanied by vigorous droplet ejection (Figure S7b). The droplets are rapidly propelled away from the heated surface and bounce up. This observation indicates that the interaction between the droplet and the laser-induced microstructures belongs to the violent transition boiling state.