Low friction droplet transportation on a substrate with a selective Leidenfrost effect

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ABSTRACT

An energy saving Leidenfrost levitation method is introduced to transport micro-droplets with virtually frictionless contact between the liquid and solid substrate. By micro-engineering the heating units, selective areas of the whole substrate can be electro-thermally activated. A droplet can be levitated as a result of the Leidenfrost effect, and further transported when the substrate is tilted slightly. The selective electro-heating produces a uniform temperature distribution on the heating units within 1 s, in response to a triggering voltage. Alongside these experimental observations, finite element simulations are conducted to understand the to understand the role of the substrate thermal conductivity on the temperature profile of the selectively heated substrate,. We also generate phase diagrams to verify the Leidenfrost regime for different substrate materials. Finally, we demonstrate the possibility of controlling low friction high speed droplet transportation (~ 65 mm/s) when the substrate is tilted (~ 7 °) by structurally designing the substrate. This work establishes the basis for an entirely new approach to droplet microfluidics.

INTRODUCTION

Transporting droplets in a controllable and energy efficient manner could have a significant impact on several engineering applications, such as low drag liquid transportation[1,2], water collection[3,4], and advanced microfluidic devices[5]. Based on a theoretical understanding of the wetting of surfaces, common approaches usually focus on creating a surface with designed physical/chemical features, e.g. hierarchical micro/nanostructured surfaces[6,7], chemical gradients[8,9], or slippery surfaces created by infusing low surface tension lubricant into microstructures that yield directional motion of water droplets when tilted at a low angle[10-12]. Notably, some interesting attempts have demonstrated liquid transportation efficiency using such techniques: for example, Chaudhury and Whitesides achieved an average velocity of 1−2 mm/s for droplet transportation on a silicon wafer possessing a gradient in wettability[13], Ghosh *et al.* employed extreme wettability patterns to achieve a flow rate of up to 300 mm/s[14], and Lv *et al*. achieved a maximum speed of 420 mm/s for droplet transportation in a microfluidic system[15]. Approaches taken to date have been based on the liquid-solid contact, where the droplet motion will more or less be affected by the friction or dragging effect induced by local surface roughness, dimensional confinement, as well as the non-uniformity of the surface wettability.

The Leidenfrost phenomenon (Figure 1a), first discovered in 1756[16], describes a meta-stable state of a droplet on a substrate heated significantly above the boiling point of the liquid. In this state, the droplet is levitated by an instantaneously generated vapor layer (~ 100-200 μm) caused by the initial contact of the droplet with the substrate[17-21]. The levitation yields a virtually frictionless contact between the droplet and substrate[21-23], therefore playing a key role in drag reduction for the liquid flow[24,25]. Moreover, the vapor layer acts as a thermal insulator preventing rapid droplet evaporation despite the high temperature of the substrate. Recent developments show some attempts to control the droplet motion based on the Leidenfrost effect levitation by employing ratcheted and other patterned substrates[21,26-29], magnetic fields[30], electric fields[31] and acoustic radiation pressure[32]. However, the actuation of the Leidenfrost effect for a droplet has thus far involved heating the entire substrate, which limits downstream applications due to the extreme substrate temperature condition. Despite recent work to reduce the transition temperature[24,28], the high energy consumption remains due to heating the substrate globally, rather than the localized area which supports the droplet.

In this study, we trigger Leidenfrost levitation of a droplet by the application of a voltage to micron-scaled serpentine shaped heating arrays, which cover the substrate in a selective manner.

In addition to initializing the levitation of droplets of three different liquids via selective heating of substrate areas, we also show that the droplet transportation can be actuated and controlled by designing heating array patterns, along with tilting of the substrate (5−10 °). The proposed strategy of selective heating could significantly reduce the energy input needed to actuate the Leidenfrost effect, and also offer a control mechanism for droplet motion by locally controlling the designed heating array. By combining our approach with surface relief patterns, precise directional control and self-propulsion can be achieved without the need to tilt a surface[33]. It also enables the possible integration of levitation of droplets into micro-systems as a new type of on-chip platform.

EXPERIMENTAL

Micro-fabrication: Thin film resistors were fabricated on different substrates, i.e. borosilicate glass and silicon (with oxidation layer) wafers. After cleaning the substrate, a thin film metal layer (Cr/Au = ~10 nm/~100 nm in thicknesses) was coated on the substrate via electron beam evaporation. The resistor patterns were photolithographically transferred into the metal layer by spinning Megaposit SPR-350 photoresist, which was exposed to UV light in an EVG mask aligner and then developed in Microposit MF-319 developer for 90 s. The excess metal out of the photoresist's protection was removed using selective gold (4:1:8 $KI:I_2:H_2O$) and chromium (7:34:1 $Ce(NH_4)_2(NO_3)_6:HNO_3:H_2O$) wet etches, leaving the required resistor patterns with varying distance between consecutive lines.

Leidenfrost Levitation Activation and Measurements: Each wafer was selectively covered by 4 two dimensional arrays of devices, and electrical contact pads for each array were designed to enable independent control of the heating arrays. A customized rig was assembled with a stainless steel stage on an $x\ y\ z\ \theta$ manipulator, to assist the characterization. In order to maintain a uniform temperature within the stage, an mbed-controlled Peltier cooler with cooling pipes and fan was mounted on the underside of the stage to stabilize the ambient condition. Spring loaded electrical contacts were then used to pass a current through each array in turn. To verify the effectiveness of the Leidenfrost Levitation, we tested three different liquids, isopropanol (surface tension ~ 20 mN/m), acetone (surface tension ~ 28 mN/m), and deionized water (surface tension ~ 72 mN/m). All liquids produced similar results, once heated beyond their respective Leidenfrost transition temperatures. In this paper, we present the summarized results for IPA as a typical case for the lowest surface tension liquid, which is the most difficult liquid when attempting to use materials techniques to create a super-liquid-repellent state. A 1000x USB optical microscope was used to observe the arrays, and a FLIR A40 thermal camera was used to observe the temperature profile of the substrate. Different powers were applied to each heated array in turn, and the array was left for 1 minute for the temperature to equalize.

Simulation: A 'unit cell' device was also modelled in COMSOL Multiphysics software, which also included the substrate, stage and effects of the Peltier cooling as a boundary condition. The COMSOL model involves parameterized substrate gaps between consecutive unit cells, and so all heating ratios can be simulated automatically. The substrate either side of the unit cell is related to the heated ratio, with the end of the substrate being the halfway point between two unit cells, which would be the coldest point in the array and therefore the region most likely to cause a collapse in the Leidenfrost vapor layer. All of the solid vertical boundaries in this model have a symmetry condition, whereas the air has an outflow condition. Finally, the bottom of the stainless steel domain has a fixed temperature boundary condition (21 °C), to simulate the Peltier cooler placed underneath the stainless steel stage.

RESULTS AND DISCUSSION

In our experiments, the concept of selective electro-thermally actuating Leidenfrost levitation is achieved by engineering millimeter-scale heating units on the substrate. The heating units covering the substrate are intended to create a uniform distribution of the thermal energy to trigger the Leidenfrost effect (Figure 1b), but in a selective manner only where this is needed to levitate a droplet. Different substrates (borosilicate glass and silicon) were used to determine the effect of the substrate thermal conductivity on the power needed for the Leidenfrost effect to occur. Since the thermal conductivities for borosilicate and silicon are 1.14 W/mK and 1480 W/mK respectively, these choices provide over three orders of magnitude difference with this parameter[32]. The silicon substrates were electrically insulated via a 100 nm thick silicon dioxide layer, grown by furnace oxidation, between the substrate and the heating array layer. In preliminary experiments we confirmed that the Leidenfrost effect could be achieved with droplets of isopropanol (IPA), acetone, and deionized water. Once the Leidenfrost effect was triggered, virtually frictionless liquid transportation was expected on a pre-tilted substrate.

We first consider a substrate patterned with relatively large heating units arranged in 2.5 mm width bands. After tilting the substrate by ~ 7 °, the levitated isopropanol (IPA) droplet (~ 40 μL) is transported across a distance of 17.5 mm on the substrate in 0.27 seconds, i.e. ~ 65 mm/s, indicating very low friction (Figure 1c). The Leidenfrost effect usually represents a meta-stable state of droplet when it comes into contact with a surface that is significantly hotter than the liquid's boiling point. Typically, a vapor layer will be initialized to support, and so yield a longer lifetime, of the droplet, and enable a virtually frictionless contact between the drop and the substrate. The droplet transportation supported by an electro-thermally actuated Leidenfrost effect is demonstrated for the first time in this report.

The actuation of Leidenfrost effect was further investigated by designing and patterning micron-scaled heating arrays onto substrates. To quantitatively evaluate the electro-heating actuation of the Leidenfrost effect, we designed the serpentine-shaped repeating 'unit cell' (Figure 2a) to further reduce the surface coverage to 62.5% of the overall area of the heated region. A defined geometrical parameter, the heating ratio, which is a 1 dimensional ratio between heated and unheated regions, where p = 40 μm and represents the width of the heated region, and the ratio is $p{:}(p + d)$, where d is the distance between consecutive unit cells, seen in the inset of Figure 2b. The total area heated for a given ratio can be calculated by multiplying the heated ratio by the serpentine unit cell coverage. For example, for a 0.2 heated ratio, the total heated percentage would be 20% multiplied by 62.5%, which is 12.5% of the area being heated. A typical plot of evaporation time versus input power on a heated 2D array of unit cells, shown in Figure 2b, is similar to previous results using more conventional methods (e.g. hotplate heated devices) (Figure S-1). In Figure 2b, the Leidenfrost transition (dotted line) is monitored by recording the evaporation time of a droplet, t_{eva}, as a function of the power input for a 0.5 heated ratio array. The dotted line of 21.8 W is the typical power value needed to boil the IPA droplet (for these experiments, a 20 μL volume was used) for this heated ratio, whereby the IPA touches the hot surface and evaporates due to the high temperature of the surface below it. Above this power, the transition regime denotes a region where the substrate is hot enough to begin creating a localized vapor underneath the droplet, but the temperature is not yet high enough to do this in a stable way, and the vapor layer is not thick enough to maintain a levitating droplet. In the stable Leidenfrost regime (right of the dotted line), the vapor can maintain a stable levitated state for the droplet. The determination of transition regime under selectively heated electrical actuation reveals a strong similarity to that from global heating of the substrate.

In contrast to heating a substrate globally, selective heating to create discontinuously heated fields across the in-plane area of the substrate should reduce the energy input. To understand how the thermal energy distributes across the substrate, we performed surface thermal analysis using COMSOL Multiphysics. The qualitative analysis (Figure 3a) first considers a serpentine 'unit cell' resistor (0.2 heated ratio) on a borosilicate glass substrate with a voltage applied to show the temperature profile of the single unit cell above the Leidenfrost transition temperature. The heat created as a result is then dissipated through the substrate, the stage underneath and also the air above the resistor. As can be seen in Figure 3a, the temperature difference across a distance of 100 μm, from the center of the heat to half way between two unit cells (or a heated ratio of 0.2) is as high as 20 °C for the borosilicate glass substrate.

As a result of this temperature difference, it would be expected that the resistors with a lower heated ratio (or a larger gap between them) would have to be heated to a higher temperature than required, in order to get the coolest part of the array to still be hot enough for the Leidenfrost effect to occur. To prove this, we further simulate the unit cells, with three adjacent serpentine units shown here on the same substrate (Figure 3b) with a voltage applied on each unit cell (heated ratio = 0.5), and demonstrate a more uniform distribution of the temperature (the difference of temperature is less than 5 °C). In this case, the voltage required is lower than for the unit cell shown in Figure 3a. We note that the current flow through the serpentine-shaped unit cell is non-uniform as a result of the structure's geometry, as can be seen in Figure S-2, and the subsequent thermal stress localization could potentially lead to mechanical failure. However, no failures occurred in our experiment and this may have been because the localized strain energy was likely absorbed by the in-plane structural expansion and the chromium adhesion layer.

Using the selectively heated substrate with repeated unit cell (heated ratio = 0.5), we next plot the phase diagrams to describe the meta-stable state of the heated IPA droplet on a glass substrate (Figure 3c) and a silicon substrate (Figure 3d). Experimental data are compared with the COMSOL simulation results. The Leidenfrost state is shown in the diagram and the anticipated trend of reducing power being needed to initiate the Leidenfrost effect when increasing the heating ratio is observed. There is good agreement between the experimental and simulated results, where the model assumes a Leidenfrost temperature of 220 °C, which is a reasonable value for IPA which has a boiling point of 82.4 °C[34].

As the selectivity of the Leidenfrost effect is via voltage actuated heating units on the substrate, the thermal conductivity of the latter will have a direct influence on the results. Two substrate materials with a large difference on thermal conductivity were employed to verify this impact, i.e. borosilicate glass (1.14 W/mK) and silicon (1480 W/mK) at 300 K[32]. As can be seen in Figure 3d, the results for a silicon substrate still follow the decreasing trend with increasing heated ratio. However, a greater power per line is required than for the borosilicate substrates to achieve the Leidenfrost effect, because the heat is dissipated through the substrate more readily, rather than remaining in the vicinity of the serpentine-shaped unit cells to form the uniform in-plane temperature profile. Therefore, borosilicate is a more preferable substrate material for this experiment, as it could create a uniform temperature profile more effectively on the surface of the substrate, owing to its low thermal conductivity.

Finally, we demonstrate the possibility of controlling droplet transportation by taking advantage of using a selectively heated substrate (Figure S-3). A substrate with four separate blocks of 0.5 ratio heating arrays, each of which could be individually switched in sequence was created, as seen in Figure 4a and 4b. The first, second and fourth arrays were switched on for 0.1 s in turn, with a 0.1 s time gap between each one being switched on. Therefore, the substrate was selectively heated in the micro-scale (due to the heated ratio), the macro scale (the four arrays being heated individually) and in time (the arrays being sequenced individually). Each array was activated for an eighth of a cycle. The third array was left disconnected. A schematic of the controlled droplet transportation shows the droplet to the target zone (un-activated region) possessing the disconnected heating array in a short time. Experimental images (Figure 4c and 4d) indicate rapid droplet movements from both directions to region #3, at a comparable speed to that witnessed in Figure 1c, thus implying rapid, virtually frictionless transport. This concept also enables a new strategy of targeted delivery of a droplet by configuring the substrate to form, on demand, localized frictionless levitation layers, which could be of considerable interest to scientists and researchers in micro-fluidic systems, chemical engineering, biological engineering and other related areas.

CONCLUSION

In conclusion, we have demonstrated the electro-heating actuation of the Leidenfrost effect for three different liquids by applying voltages to heating units which selectively cover a substrate. This approach provides rapidly switchable and highly targeted transportation of droplet, according to the design of the geometry and layout of the heating array on the substrate. By selectively heating a sample to produce the Leidenfrost effect in a small area where needed, rather than using a hot plate to heat the entire surface, the same effect can be achieved but with a much lower energy requirement. It also provides the potential for easy integration to be part of an on-chip device with an electrical triggering mechanism. Moreover, further energy efficiency could be achieved by using a feedback control system to drive and control the direction of motion of droplets with actuation only of those heating units in the instantaneous locality of the levitated droplet.

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SUPPORTING INFORMATION

- **Figure S-1.** The Leidenfrost transition graph for a sample on a hotplate
- **Figure S-2**. COMSOL image of flow of current through serpentine resistor on a borosilicate substrate.
- **Movie S-3.** Video showing targeted transportation of a droplet upon selective heated substrate. We show the instant droplet transportations from both directions to expected destination when we applied a voltage of 60 V to the settings in Figure 4a and 4b. The droplet travels within 1 second and stopped at the non-activated area (#3).

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FIGURES

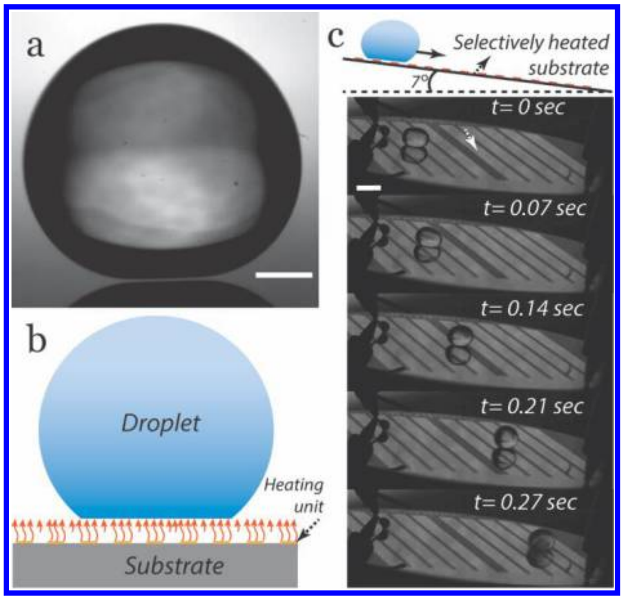

Figure 1. Initializing the Leidenfrost effect by creating localized Joule heating areas with selective coverage of heating units on a substrate. (a) A side view image of a traditional Leidenfrost levitated water droplet on a heated silicon wafer surface (~ 300 °C); the scale bar is 500 µm. (b) Schematic illustration of Leidenfrost levitation on a substrate selectively covered by metal heating units. (c) The concept is examined through virtually frictionless transportation of an IPA droplet (~ 40 µL), supported by the levitation vapor layer. The substrate is selectively covered by heating units in bands of width 2.5 mm (indicated by white the arrow) indicating droplet transportation speeds of ~ 65 mm s^{-1}; the substrate is tilted by 7 ^{o} and the scale bar is 2 mm.

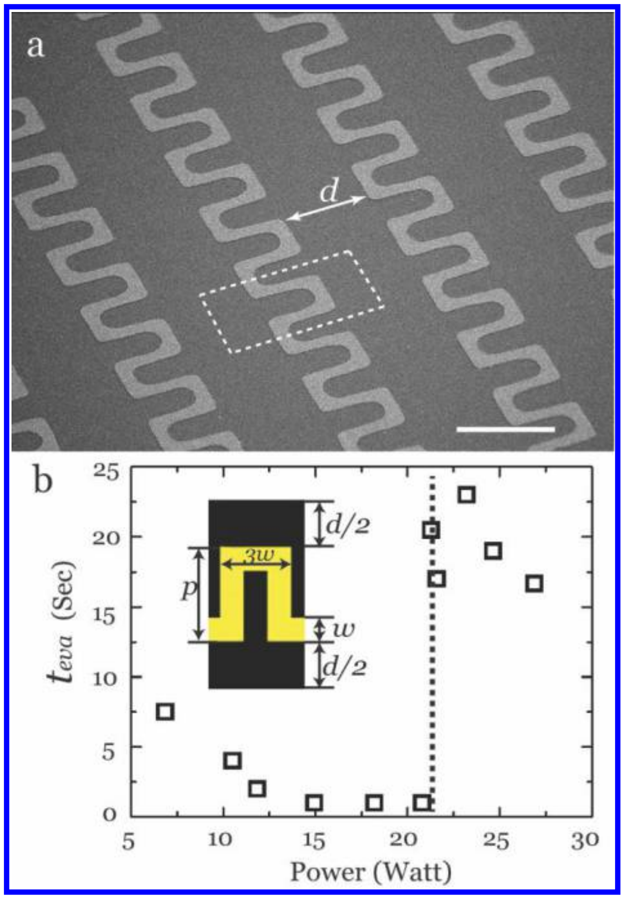

Figure 2. Micro-pattern of gold heating units and detection of the critical Leidenfrost transition. (a) SEM image of micro-patterned chromium/gold heating array using serpentine patterns defined by an electrode width w = 10 μm, gap distance d, and electrode structure width p = 40 μm. (b) The Leidenfrost transition (dotted line) is monitored by recording the evaporation time of a droplet, t_{eva}, as a function of the power input. The inset shows the unit cell design within a serpentine electrode for selective heating of the substrate (the heated area is the lighter area), where the overall unit area is shown by the dashed box in (a).

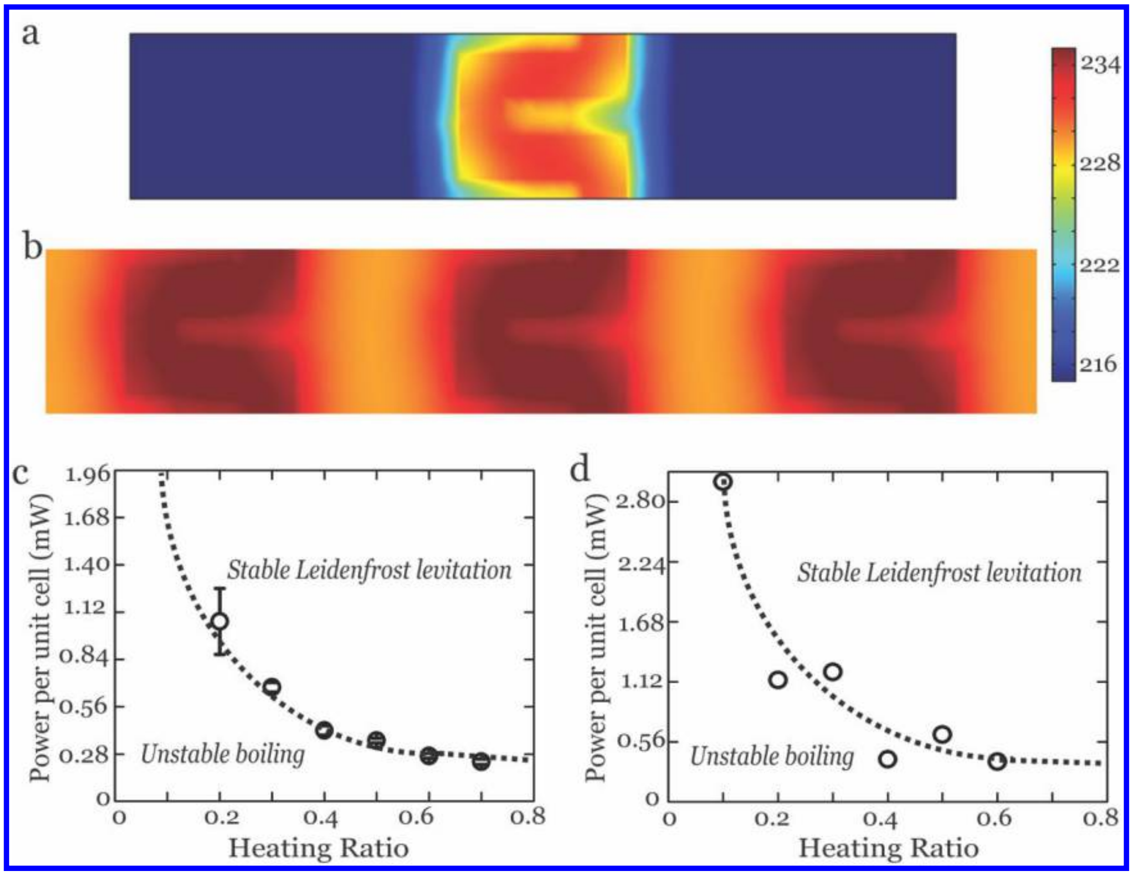

Figure 3. Selective Leidenfrost mechanism and phase diagrams. (a) Temperature profile of surface for the electro-heating of single unit cell with a 0.2 heated ratio. The COMSOL simulation in (b) reveals a uniform distribution of the temperature (in degrees Celsius) across the unit cells within three adjacent serpentine units (a heated ratio of 0.5). The simulation is based on 363 serpentine unit cells in series, which are referred to as a line, and a number of these lines are connected in parallel to form a complete heating array. The applied voltage is then varied to provide an equivalent power per cell required for the Leidenfrost effect to occur. The selective Leidenfrost effect using this pattern is described by phase diagrams on (c) a glass substrate and (d) a silicon substrate. The dotted lines represent the simulation results and the symbols represent the experimental results, respectively.

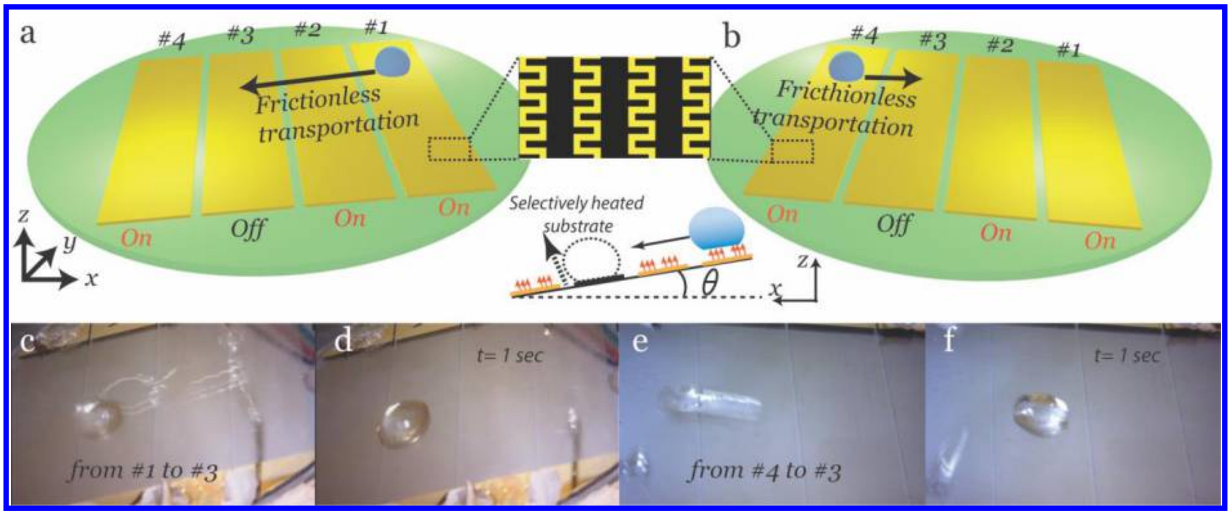

Figure 4. Demonstration of droplet transportation using programmable activation of localized heating units. (a-b) Schematic illustrations of experimental arrangements with example designs which selectively cover a substrate with four micro-patterned heating arrays (numbered #1 to #4 from right to left); each complete unit consists of serpentine-patterned unit cells as illustrated by Figure 2a. The third unit is non-activated to give an 'off' state while the others are activated into an 'on state'. The whole wafer is tilted at a small angle of 10 ^{o} in (a) and -10 ^{o} in (b), as shown in the inset side view schematic. The droplet travels downslope across regions #1 and #2 with almost no friction due to the Leidenfrost effect, but then stops when it reaches the non-activated region #3; droplet transport is also stopped on the non-activated region when the substrate tilt is reversed (b). (c) Momentary snap shot of the droplet transportation shows the droplet advances from region #1 to region #3 with the setting in (a), and finally stopped at the non-activated region #3 (d). (e-f) Experimental images for droplet transport records the rapid virtually frictionless droplet transport from region #4 to region #3 with the setting (b).

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