Shaping high-speed Marangoni flow in liquid films by microscale perturbations in surface temperature

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The authors show that a variety of controlled flow patterns, including toroidal cells and surface doublets, can be generated in 80–400 μm thick liquid films by placing scanning microscopy probes with integrated heaters just above the surface (<400 μm separation). The probes project sharp temperature gradients on the liquid surface which drive Marangoni flow. Flow velocities approaching 3000 μm/s are experimentally demonstrated on length scales of 20–200 μm with <20 mW input power. For liquids such as water and oil, in which the surface tension coefficient is ≈0.2 mN/m K, flows >1000 μm/s can be accomplished with surface temperature perturbations <1 °C. This technique enables microfluidic manipulation on unpatterned substrates. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2430777]

Microfluidic actuation methods exploiting surface forces (which scale favorably at small length scales) are being actively researched for use in miniaturized biochemical assays and other applications. The actuation of microdroplets, for example, typically utilizes interfacial forces generated by electrowetting,[1,2] substrate temperature gradients,[3,4] chemical gradients,[5,6] and dielectrophoresis.[7] Nevertheless, most actuation techniques utilize prefabricated (and frequently complex) microchips that are typically assay specific. We show that microfluidic actuation of films, droplets, and suspensions can be achieved by the Marangoni effect, simply by creating temperature perturbations on the free surface of the liquid sample using heated or cooled scanning microscopy probes (Fig. 1). This letter focuses on toroidal flows and doublets (Fig. 2), which are two of the most elementary flow patterns that can be generated with a simple, pointlike heater geometry. Experiments show how these flow patterns can also be used for trapping and agglomerating particles and droplets. In addition to providing a method of instrumentation, these results illuminate the dramatic impact of surface temperature perturbations on microscale flow.

The microfabricated probes used in the majority of the following experiments to create the thermal perturbations are thin film cantilevers similar to atomic force microscopy probes, except that they contain an integrated joule heater near the tip.[8] The cantilever, with an orthogonal tip, extends from the edge of an upturned silicon chip. It is made with thermally insulative polyimide; an embedded thin film of gold, which covers the tip, also serves as a resistive heater. A typical probe is 360 μm long, 120 μm wide, 3 μm thick, and has resistance ranging from 20 to 50 Ω. The tip temperature can reach nearly 50 °C with <5 mW input power because the low thermal conductivity of the polyimide suppresses conductive loss.

A probe tip placed within about 400 μm of the liquid creates a localized increase in the surface temperature. The consequent reduction in surface tension creates a shear stress that is oriented radially outward, driving fluid flow away from the hot spot on the surface. The continuity is maintained by inward flux beneath the surface. Together, the surface and subsurface currents form a recirculating convection cell which is axisymmetric about the heat source [Fig. 2(a)]. In a typical toroidal cell, the radial stress and resultant fluid velocities are proportional to the surface temperature gradient, according to the surface stress condition boundary condition[9]

$$\Gamma_r = \mu \frac{dV_r}{dn} = -\frac{d\sigma}{dT}\frac{dT}{dr}, \qquad (1)$$

where Γ_r is the radial shear stress, μ is the fluid viscosity, dV_r/dn is the gradient of radial velocity perpendicular to the surface, $d\sigma/dT$ is the temperature coefficient of surface tension, and dT/dr is the radial temperature gradient on the fluid surface. The speed and geometry of the flow can be controlled by the gap between the sample and the probe and the power supplied to it. For example, Fig. 3(a) shows measured flow velocities as a function of probe power obtained

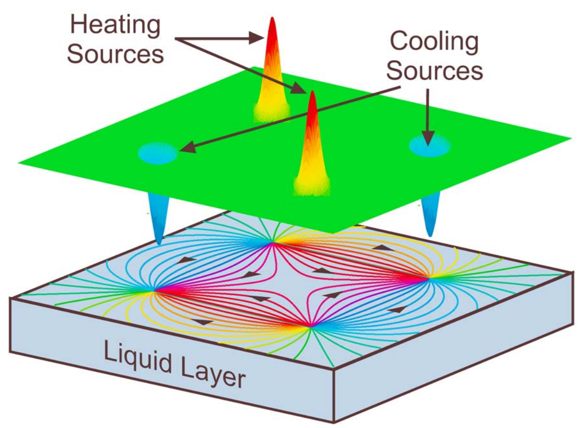

FIG. 1. (Color online) Controlling microfluidic flow with Marangoni convection. Thermal sources suspended above the surface of a thin liquid film provide arbitrary, patterned heat fluxes to the liquid surface, resulting in controlled surface tension gradients. Point heating sources causes flow away from the heated surface, whereas point cooling sources cause inward flow. Heat fluxes can be designed to produce complex flow patterns, as illustrated here. Fast flow velocities (>3 mm/s) can be achieved with small increases in surface temperature (<3 °C) when this technique is applied at small length scales.

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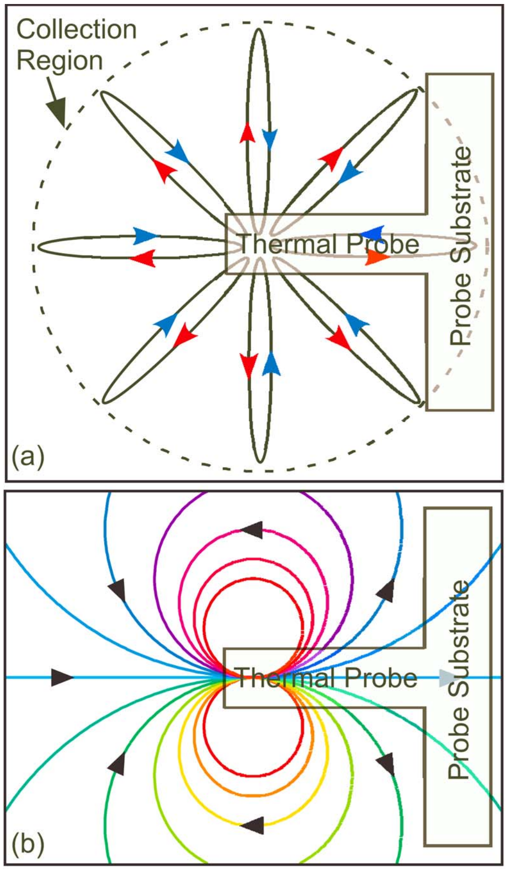

FIG. 2. (Color online) Schematics of toroidal and doublet flows. (a) Top view of toroidal flow, which occurs when the probe is suspended above a thin oil layer. Surface flows (red arrows) are directed outward, and subsurface flows (blue arrows) are directed inward. (b) Top view of doublet flow, which generally occurs in water and is a two dimensional surface flow. The positioning of the probe relative to the flow is shown.

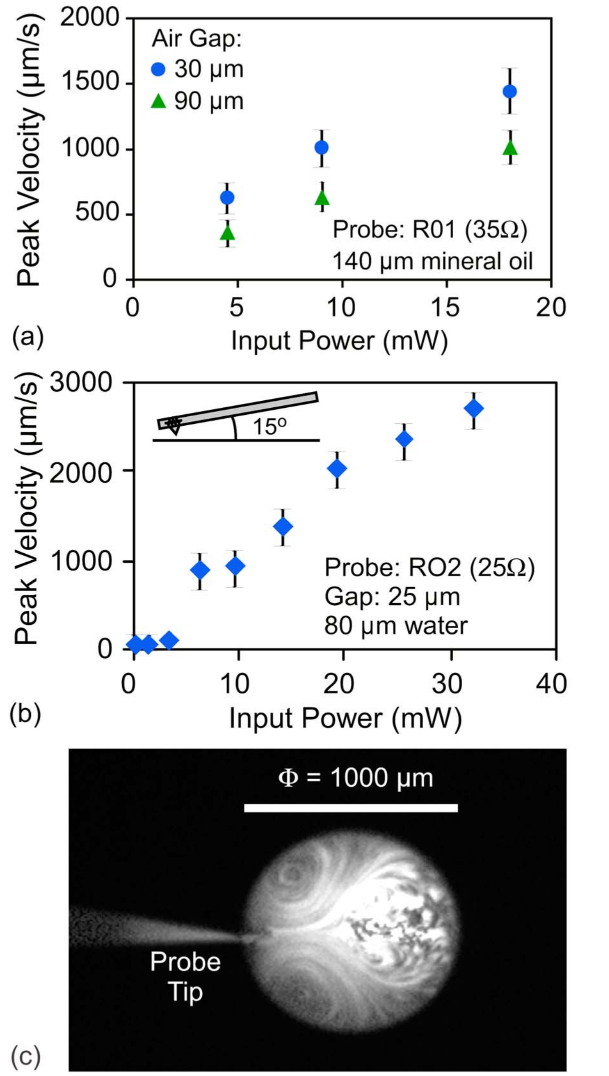

FIG. 3. (Color online) Experimental data. (a) Toroidal flow velocities as a function of input power to the probe. Power to the probe was ramped while the probe-liquid separation was fixed at 60 and 90 μm. Flow velocities were recorded with a charge coupled device camera. (b) Doublet flow velocities as a function of input power when the probe is held at an 80 μm separation. (c) Fluorescent images of the doublet flow within a 1 mm droplet when a heated probe tip is held next to a droplet submerged in oil.

on a 140 μm thick layer of mineral oil. This result is consistent with Eq. (1). The radius of the convection cell depends primarily on the thickness of the oil layer. In thin fluid layers (<150 μm), the solid substrate below the liquid film absorbs the heat, diminishing the lateral flow of heat. In addition, the increased viscous drag impedes the opposing surface and subsurface flows. As a result, the radial extent of the flow diminishes. Simulations indicate that for fluids with a surface tension coefficient >0.2 mN/m K, such as various oils, it is possible to obtain fluid velocities >1000 μm/s while the surface temperature rise is <1 °C. The small temperature increase required makes this technique a candidate for many samples and assays. (A cold probe, which actively cools a localized region on the fluid surface, generates Marangoni flows in the reverse direction.) These flow patterns offer control and usability that is not possible with self-organized cellular flow ensembles created by substrate heating.[10,11]

Aqueous samples exhibit high speed doublet flow patterns rather than toroids. Doublets are associated with linear thermal gradients created by the polyimide probes. The silicon chip to which each probe is attached presents a heat sink, creating a surface temperature gradient. (Simulations and other evidence[12] suggest that evaporative cooling may additionally play a role in such cases.) Hence, the angle at which the probe is held above the surface (i.e., the attack angle) has an impact on these flow patterns. For example, with a 15° attack angle above an aqueous film of 80 μm thickness, linear velocities of up to 3000 μm/s can be achieved in the center streamlines. Changing the attack angle can even reverse the flow. The fluid velocities can be controlled by adjusting the gap between the probe and liquid, or by adjusting the power to the probe [Fig. 3(b)]. Rotational velocities in the doublet vortices approach 1300 rpm, indicating a potential for use in laminar mixing, which is often difficult to achieve at high speeds at the microscale.[13,14]

Marangoni flows could become a potentially useful tool in microdroplet-based chemical analysis sytems, where submillimeter-sized droplets act as fluid compartments, and reactions can be initiated by merging the droplets. Since the heat sources do not make contact with the liquid, the chance of sample loss or sample contamination is reduced. The recirculating nature of toroidal flows makes them suitable for collecting and manipulating droplets. Doublet flows, on the other hand, could be used for mixing within individual droplets. Figure 3(c) shows the streamlines of high-speed vortices generated within a 1 mm droplet.

We conclude that thermal perturbations on fluid surfaces can control Marangoni flow at the microscale, and it can effectively obtain fast velocities with relatively low power and voltages. These perturbations could be extended to multiple line and point sources as well as arbitrary two dimensional arrays in order to produce geometrically complex flow patterns in fluid films and pools.

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