

Optical microfluidics

Cite as: Appl. Phys. Lett. **85**, 2658 (2004); <https://doi.org/10.1063/1.1797538>

Submitted: 17 February 2004 . Accepted: 27 July 2004 . Published Online: 28 September 2004

K. T. Kotz, K. A. Noble, and G. W. Faris



View Online



Export Citation

ARTICLES YOU MAY BE INTERESTED IN

[Thermocapillary manipulation of droplets using holographic beam shaping: Microfluidic pin ball](#)

Applied Physics Letters **93**, 034107 (2008); <https://doi.org/10.1063/1.2952374>

[Marangoni flow at droplet interfaces: Three-dimensional solution and applications](#)

Physics of Fluids **28**, 012106 (2016); <https://doi.org/10.1063/1.4939212>

[Electrowetting-based actuation of liquid droplets for microfluidic applications](#)

Applied Physics Letters **77**, 1725 (2000); <https://doi.org/10.1063/1.1308534>

Lock-in Amplifiers
up to 600 MHz



Optical microfluidics

K. T. Kotz, K. A. Noble, and G. W. Faris^{a)}

Molecular Physics Laboratory, SRI International, 333 Ravenswood Avenue, Menlo Park, California 94025

(Received 17 February 2004; accepted 27 July 2004)

We present a method for the control of small droplets based on the thermal Marangoni effect using laser heating. With this approach, droplets covering five orders of magnitude in volume ($\sim 1.7 \mu\text{L}$ to 14 pL), immersed in decanol, were moved on an unmodified polystyrene surface, with speeds of up to 3 mm/s . When two droplets were brought into contact, they spontaneously fused and rapidly mixed in less than 33 ms . This optically addressed microfluidic approach has many advantages for microfluidic transport, including exceptional reconfigurability, low intersample contamination, large volume range, extremely simple substrates, no electrical connections, and ready scaling to large arrays. © 2004 American Institute of Physics. [DOI: 10.1063/1.1797538]

Much of the work on microfluidics to date has involved production of patterned fluidic circuits,^{1,2} a relatively direct analogy with microelectronics. An alternative approach involves movement of individual droplets upon a surface. While fluidic circuitry is the microscopic equivalent of tubing or pipes, droplet motion is a closer analogy to the individual droplet delivery obtained with pipettes, the current mainstay of the macroscale chemistry and biology laboratory. Droplet-based techniques thus provide an alternative paradigm to current lab-on-a-chip technologies for high throughput applications in chemistry and biology using small volumes and large numbers of samples.

Surface forces are the primary challenge for moving droplets on a solid surface. The strength of adhesion is indicated in part by the contact angle. Figure 1 shows a 1.8-mm -diameter water droplet on a polystyrene surface immersed in decanol. The contact angle between the droplet and the surface (θ_e) is close to 180° , indicating relatively little surface adhesion. When a force is applied to the droplet, the symmetry between the contact angles is broken. The difference between the advancing and receding contact angles, or contact angle hysteresis, leads to a net force counteracting the applied force. The force required to move the droplet scales with the contact perimeter and the contact angle hysteresis.³ Although we have moved droplets using radiation pressure forces (optical traps or optical tweezers),⁴ we find it is preferable to use surface tension forces to move the droplets.

Surface tension and surface energy generally drop as temperature is increased.⁵ If a thermal gradient is applied along a droplet, the droplet moves toward the colder region to minimize the total surface energy, an effect called the thermal Marangoni effect. This effect can lead to formation of fingers extending from a pool of liquid onto a cooler surface. Control of these fingers has been achieved using regions of varying surface energy⁶ or, recently, optical heating to produce patterned variation in surface energy.⁷ Thermal gradients have also been used to produce rapid motion of droplets in combination with a chemically-induced surface energy gradient,⁸ to flow droplets in a channel,⁹ and to manipulate droplets on a surface.¹⁰ Similar surface tension imbalances generated with electric field gradients have also

been used to move droplets.^{11,12} Electric field gradients may also be used to move droplets through dielectrophoresis.^{13,14} Light forces have been used for fluidic control using photoisomerization to change the surface energy¹⁵ or through optical traps either through direct manipulation of liposomes^{16,17} or through optical manipulation of colloidal microspheres.¹⁸ We show how a laser beam may be used to move droplets using the thermal Marangoni effect.

Aqueous droplets were deposited in an organic phase of 1-decanol on top of a standard polystyrene petri dish by two different methods. The first was by dragging the tip of a 34-gauge needle ($100 \mu\text{m}$ inner diameter) across the bottom of the petri dish. The second was through the direct ejection of small droplets into the organic phase from a standard ink-jet print head (HP 51625A). We used the former method to produce droplets ranging in size from $40\text{--}1500 \mu\text{m}$ in diameter, while the latter method was capable of reproducibly generating aqueous drops on demand with diameters of $30\text{--}40 \mu\text{m}$.

We move the droplets via an induced surface energy gradient, shown schematically in Fig. 2. We focus $10\text{--}200 \text{ mW}$ of green light from an argon ion laser onto the imaging plane of an inverted microscope, controlling the position of the focus in the plane with a motorized mirror. The droplets contain an aqueous dye (FD&C Red No. 40 and Red No. 3, McCormick & Co. Inc.) that is diluted to a typical absor-

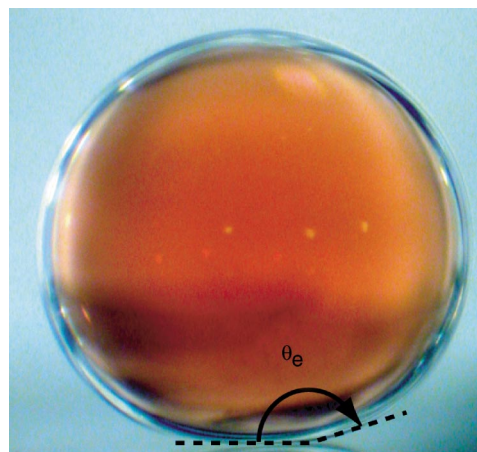


FIG. 1. (Color online). Photograph of the equilibrium contact angle (θ_e) for a drop of water in decanol on a polystyrene surface.

^{a)} Author to whom correspondence should be addressed; electronic mail: gregory.faris@sri.com

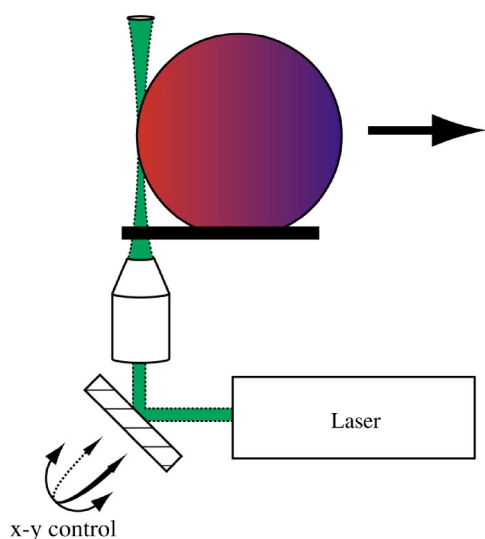


FIG. 2. (Color online). Schematic diagram of the experimental apparatus used to move the water droplets.

bance of 2/cm at the laser wavelength. The absorption forms a temperature gradient across the drop (Fig. 2), which in turn induces a surface energy gradient on the droplet surface via the Marangoni effect that is sufficient to move the droplets. Unlike prior thermal Marangoni-driven fluidics,^{7–9} the surface tension forces here arise at a fluid/fluid boundary^{19,20} rather than at the fluid/solid boundary. It should be noted that the decanol influences the water/surface contact angle and also acts to control the evaporation of the droplets. Although we use a dye in the droplet for this work, it is not required. An alternative would be to add a dye to the surrounding fluid, or, preferably, to use a laser that is selectively absorbed by either the droplet or the surrounding fluid so that no dye is required at all.

Using this simple method, we have been able to move droplets as large as 1.5 mm and as small as 30 μm in diameter. This corresponds to a range of volumes that spans five orders of magnitude ($\sim 1.7 \mu\text{L}$ to 14 pL). The speed of droplet movement was limited by the slew rate of the motorized mirror mount that steers the laser beam. By physically translating the stage, however, we moved the droplets at speeds of up to approximately 3 mm/s, which corresponds to 15 diameters/s for a 200- μm droplet. By scaling observations of gravity-driven motion of 3- μL droplets when the substrate is tilted, we estimate the force required to overcome contact angle hysteresis for a 100- μm -diameter droplet is approximately 10 nN.³ Calculating the Stokes (viscous) drag on a sphere including the Faxen wall correction,^{20,21} we find a force of roughly 100 nN is required to move a 100- μm -diameter droplet at 3 mm/s, so that viscous drag dominates over forces from contact angle hysteresis in this case.

We have calculated an approximate temperature distribution in the droplet environment. When droplets are moved at the maximum velocity of 3 mm/s, the laser strikes the droplet at a distance of 1/4 of a droplet radius from the edge of the droplet, corresponding to an absorption of about 3% of the laser power for a 100- μm -diameter droplet. Solutions to the heat equation show that for an incident laser power of 100 mW, the absorbed power will produce a temperature difference of roughly 10 $^{\circ}\text{C}$ across the width of the droplet, in agreement with the gradient required to produce the 100 nN force above.^{19,20}

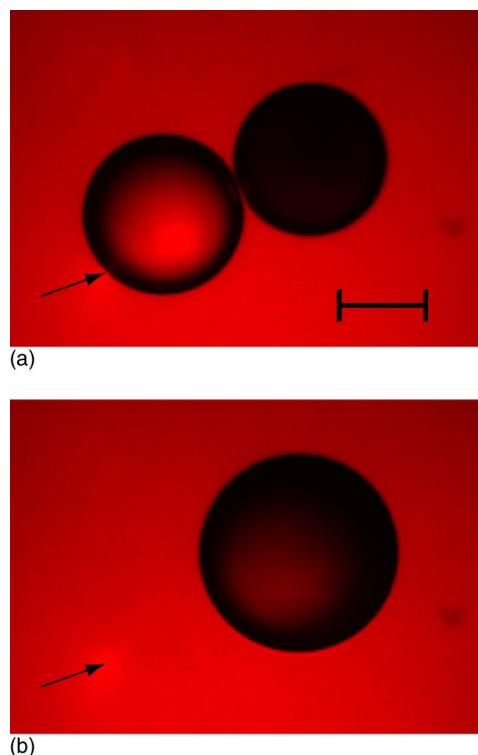


FIG. 3. (Color online). Sequence of two subsequent video frames (30 frames/s) demonstrating the fusion and mixing of two different droplets. Images are taken through a long pass (red) filter to reduce the laser intensity. (a) A dye filled droplet (187 μm diameter, lower left) has been moved by the focus of the laser (black arrow) next to a droplet containing black India ink (182 μm diameter). (b) The two combined droplets in the next video frame, 33 ms later (232 μm diameter). Droplet fusion and mixing takes place on a time scale of less than 33 ms. The scale bar in (a) represents 100 μm and applies to both panels. A video of this fusion and mixing process is available online (see Ref. 28).

We observe very interesting behavior from the small microdroplets following optical manipulation. The first is that two droplets spontaneously fuse upon contact with one another (Fig. 3). Droplet volume is conserved in each of the observed fusions, as expected. In addition, the droplets exhibit extremely rapid mixing following fusion, as shown in Fig. 3. As seen in the figure, the droplets have completely mixed in a time shorter than the time between video frames (33 ms). This spontaneous mixing appears to be a caused by the fusion process, and not by diffusion. For solutes with diffusion coefficients ranging from $5\text{--}0.5 \times 10^{-6} \text{ cm}^2/\text{s}$,²² it would take roughly 10 to 100 s to mix across 100 μm by diffusion alone. This rapid mixing is an important observation, because long mixing times often have been a challenge for microfluidics in channels.^{2,23}

The mixing in this case is driven by the change in surface energy during droplet coalescence. The fused droplet has a lower surface area, and hence lower surface energy, than the two droplets prior to fusion. The change in surface energy is largely converted to kinetic energy, causing droplet oscillation that is ultimately damped by viscosity. Using theory valid for small oscillations²⁴ and the water/decanol interfacial surface tension,²⁵ we find that 100- μm -diameter water droplets in decanol have an oscillation frequency of $\sim 1.5 \text{ kHz}$ and a damping time of $\sim 110 \mu\text{s}$.

One can define a characteristic velocity for this mixing process by equating the change in surface energy from droplet fusion with the kinetic energy of the droplet volume. In-

deed, observations of the dynamics of merging droplets with high contact angles²⁶ show contact surface velocities similar to this characteristic velocity. For 100- μm -diameter water drops in decanol, this characteristic velocity is $\sim 50\text{ cm/s}$, corresponding to a Reynolds number of about 70. The Reynolds number is a dimensionless quantity expressing the relative size of inertial and viscous forces; small Reynolds numbers are associated with difficulty in mixing. For flow over a cylinder, Reynolds numbers greater than about 2 are sufficient for formation of vortices.²⁷ Since the flow over a cylinder and the oscillations of coalescing droplets both involve direction-changing flow, it appears reasonable that vortices are formed for the droplets in our experiments. The convective motion of vortices would greatly enhance the mixing process. The internal Marangoni convection patterns¹⁹ driven by the laser beam will also contribute to mixing, although the velocities are roughly a hundred times lower.

We have also performed initial experiments generating and moving aqueous droplets at the liquid-liquid interface of decanol and fomblin (a perfluorinated silicone oil). The three-component liquid system does not exhibit contact angle hysteresis, thus making it easier to move droplets. However, the lack of contact angle hysteresis allows the droplet movement to be adversely affected by convection currents or Brownian motion (for small droplets).

In this letter we have presented an approach to microfluidics based on the optical control of thermal gradients. When compared with channel-based lab-on-a-chip techniques,^{1,2} assays based on individual droplet movement eliminate the requirement of valves and pumps; assays may be rapidly reconfigured on the fly; random access to substrate sites is easily accomplished; no on-chip "real estate" is devoted to electrical circuitry; and the substrate, generally a disposable, does not require expensive and time-consuming fabrication. In addition, our optically addressed approach holds many advantages over the existing electrically addressed droplet manipulation approaches (those using heater-based Marangoni^{9,10} or electric field gradient-based techniques¹¹⁻¹⁴) including greater reconfigurability, low intersample contamination, and ready scaling large arrays (droplet paths are not confined to moving from electrode to electrode or heater to heater), large volume range, greater substrate simplicity, and no electrical connections (which allow rapid interchange of substrates and use in harsh environments). For array manipulation, a single laser can be rapidly scanned to produce many effective beams (time sharing),¹⁸ or multiple beams can be produced from vertical cavity surface-emitting layer arrays, holograms, or spatial light modulators. Unlike liposome-based systems,^{16,17} use of this technique allows droplet production on demand; pulsed lasers are not required to fuse droplets; and, once moved, the droplets are held in place by contact angle hysteresis. The simplicity of this technique should lend itself to a number of applications, such the manipulation and study of very small fluid volumes (microfluidics and nanofluidics), the performance of very large numbers of parallel assays (high

throughput screening), stochastic studies of small samples, and the molecular analysis of large numbers of isolated cells. See EPAPS Ref. 28 for additional information.

This work was supported by the Defense Advanced Research Projects Agency and SRI International internal research funds. One of the authors (K. A. N.) was supported by the Research Experiences for Undergraduates program of the National Science Foundation. We thank K. S. Kalogerakis and S. E. Smith for assistance in assembling the scanning system and W. H. Wright for helpful discussions on optical trapping techniques.

- ¹T. Thorsen, S. J. Maerkl, and S. R. Quake, *Science* **298**, 580 (2002).
- ²J. D. Tice, H. Song, A. D. Lyon, and R. F. Ismagilov, *Langmuir* **19**, 9127 (2003).
- ³C. G. L. Furmidge, *J. Colloid Sci.* **17**, 309 (1962).
- ⁴A. Ashkin, *Proc. Natl. Acad. Sci. U.S.A.* **94**, 4853 (1997).
- ⁵A. W. Adamson and A. P. Gast, in *Physical chemistry of surfaces* (Wiley, New York, 1997), pp. 48-53.
- ⁶D. E. Kataoka and S. M. Troian, *Nature (London)* **402**, 794 (1999).
- ⁷N. Garnier, R. O. Grigoriev, and M. F. Schatz, *Phys. Rev. Lett.* **91**, 054501 (2003).
- ⁸S. Daniel, M. K. Chaudhury, and J. C. Chen, *Science* **291**, 633 (2001).
- ⁹M. A. Burns, C. H. Mastrangelo, T. S. Sammarco, F. P. Mann, J. R. Webster, and B. N. Burns, *Proc. Natl. Acad. Sci. U.S.A.* **93**, 5556 (1996).
- ¹⁰A. A. Darhuber, J. P. Valentino, J. M. Davis, S. M. Troian, and S. Wagner, *Appl. Phys. Lett.* **82**, 657 (2003).
- ¹¹M. W. Prins, W. J. Welters, and J. W. Weekamp, *Science* **291**, 277 (2001).
- ¹²S. K. Cho, H. Moon, and C.-J. Kim, *J. Microelectromech. Syst.* **12**, 70 (2003).
- ¹³T. B. Jones, M. Gunji, M. Washizu, and M. J. Feldman, *J. Appl. Phys.* **89**, 1441 (2001).
- ¹⁴J. Vykoukal, J. A. Schwartz, F. F. Becker, and P. R. C. Gascoyne, in *Micro total analysis systems 2001*, edited by J. Michael Ramsey and Albert van den Berg (Kluwer Academic, Dordrecht, 2001), pp. 72-74.
- ¹⁵K. Ichimura, S. K. Oh, and M. Nakagawa, *Science* **288**, 1624 (2000).
- ¹⁶M. Ichikawa and K. Yoshikawa, *Appl. Phys. Lett.* **79**, 4598 (2001).
- ¹⁷S. Kulin, R. Kishore, K. Helmersson, and L. Locascio, *Langmuir* **19**, 8206 (2003).
- ¹⁸A. Terray, J. Oakey, and D. W. M. Marr, *Science* **296**, 1841 (2002).
- ¹⁹N. O. Young, J. S. Goldstein, and M. J. Block, *J. Fluid Mech.* **6**, 350 (1959).
- ²⁰S. H. Chen, *J. Colloid Interface Sci.* **230**, 157 (2000).
- ²¹J. Happel and H. Brenner, in *Low Reynolds number hydrodynamics with special applications to particulate media* (Prentice-Hall, Englewood Cliffs, NJ, 1965), p. 327.
- ²²P. W. Atkins, in *Physical chemistry* (Oxford University Press, Oxford, 1986), p. 613.
- ²³A. D. Stroock, S. K. Dertinger, A. Ajdari, I. Mezic, H. A. Stone, and G. M. Whitesides, *Science* **295**, 647 (2002).
- ²⁴C. A. Miller and L. E. Scriven, *J. Fluid Mech.* **32**, 417 (1968).
- ²⁵R. Aveyard, B. J. Briscoe, and J. Chapman, *J. Chem. Soc., Faraday Trans. 1* **68**, 10 (1972).
- ²⁶A. Menchaca-Rocha, A. Martinez-Davalos, R. Nunez, S. Popinet, and S. Zaleski, *Phys. Rev. E* **63**, 046309 (2001).
- ²⁷R. L. Daugherty and J. B. Franzini, in *Fluid mechanics with engineering applications* (McGraw Hill, New York, 1977), p. 299.
- ²⁸See EPAPS Document No. E-APPLAB-85-065438 for a video showing the mixing of a dye-filled water droplet with one containing black India ink (Fig. 3 in the text). A direct link to this document may be found in the online article's HTML reference section. The document may also be reached via the EPAPS homepage (<http://www.aip.org/pubservs/epaps.html>) or from <ftp.aip.org> in the directory `/epaps/`. See the EPAPS homepage for more information.