

Actuation of Janus Emulsion Droplets via Optothermally Induced Marangoni Forces

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Micro-scale Janus emulsions represent a versatile material platform for dynamic refractive, reflective, and light-emitting optical components. Here, we present a mechanism for droplet actuation that exploits thermocapillarity. Using optically induced thermal gradients, an interfacial tension differential is generated across the surfactant-free internal capillary interface of Janus droplets. The interfacial tension differential causes droplet-internal Marangoni flows and a net torque, resulting in a predictable and controllable reorientation of the droplets. The effect can be quantitatively described with a simple model that balances gravitational and thermal torques. Occurring in small thermal gradients, these optothermally-induced Marangoni dynamics represent a promising mechanism for controlling droplet-based micro-optical components.

Many micro-fluidic technologies, including droplet-based sorting and sensing, chemical micro-reactors, and tunable fluidic micro-optics, are enabled by the precise manipulation of micro-scale droplets [1–3]. Complex droplet morphologies, such as liquid Janus particles [4] and multi-phase emulsions [5, 6], are relevant in a variety of applications, including drug delivery [7, 8], optics [9–11], biochemical sensors [12–15], and cosmetics [16]. Many of these applications rely on the ability to configure and optimize the droplet morphology. Since emulsion morphologies are usually stabilized with several surfactants, their configuration can be controlled by tuning the surfactant concentrations in the droplet medium or by dynamically modifying the surfactants’ strength, for instance through optical stimulation [5, 9]. These control mechanisms require careful tuning of the chemical environment and the design of sophisticated stimuli-responsive surfactants.

Here, we present a simple alternative for controlling emulsion droplet configuration that does not require chemical modification of the surrounding medium. The orientation of bi-phase emulsion droplets, formed from immiscible hydrocarbons and fluorocarbons, can be manipulated physically with a small temperature gradient generated with a focused near-infrared laser in the fluid medium (Fig. 1a). Droplets are observed to pan (see schematic in Fig. 1b,c) and tilt (Fig. 1d,e) as they respond to the presence and movement of the laser spot. The thermal gradient generated by the laser induces an interfacial tension gradient along the droplets’ internal surfactant-free interface (Fig. 1e). This interfacial tension gradient gives rise to Marangoni flows in-

side the droplets and a net torque, leading to droplet re-orientation. The tilt angle of the bi-phase emulsion droplets is determined by the balance of gravitational torque, which occurs due to the different densities of the constituent phases, and the torque associated with the opto-thermal Marangoni effects (Fig. 1e).

Thermal gradients are known to cause gradients in interfacial tension along a fluid-fluid interface [17–19]. Such gradients result in a net stress along the fluid interface causing Marangoni flows. Marangoni effects can also originate from chemical gradients, as observed in the famous tears of wine [20], and can cause emulsion droplets to act as self-propelling micro-swimmers, driven by spontaneous symmetry-breaking or optically stimulated surfactant reconfiguration [21–24]. The associated behavior of a single-phase droplet or bubble in a temperature gradient is well understood [25] and photothermal control of single phase droplets has been demonstrated in a number of microfluidic systems [26–30]. Theoretical studies of thermo-capillary flows in a partially engulfed multi-phase droplet show that the translation velocity of a droplet depends sensitively on its morphology and the viscosity of its constituent phases [31, 32]. In experiments, the situation is further complicated due to the presence of surfactants, which are necessary for stabilizing emulsion [33], and controlling the droplets’ morphology [5]. When subjected to a rise in temperature, surfactants may cause an increase in interfacial tension due to enhanced chemical cohesion [27], acting in opposition to conventional thermal Marangoni forces. The redistribution of surfactants on the surface will also act to mitigate any thermally-induced interfacial tension gradients [34, 35].

In our experiments, these various competing effects suppress interfacial tension gradients in the outer surfaces of the bi-phase emulsion droplets, when a thermal gradient is applied. The observed Marangoni flows and

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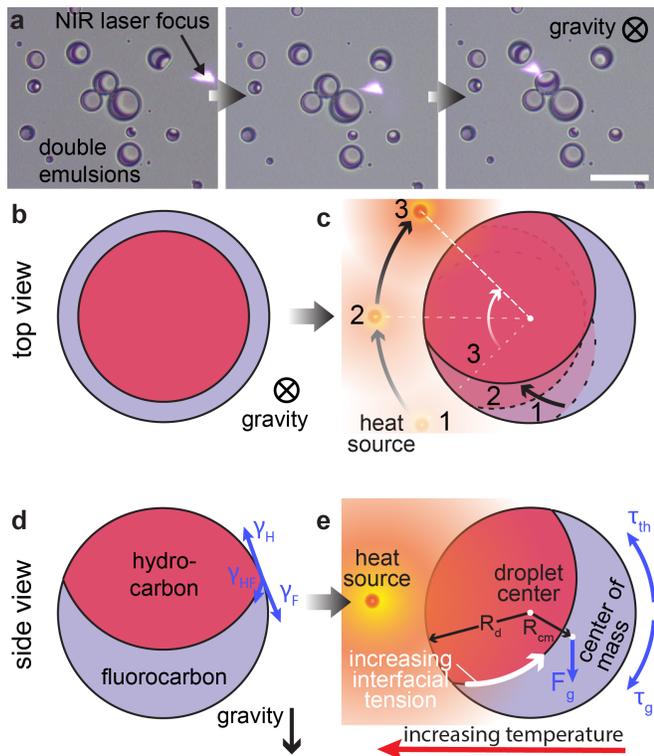


FIG. 1. **Emulsion droplets re-orient toward a laser spot, which acts as a localized heat source.** **a)** Emulsion droplets formed from heptane and perfluorohexane rotate toward the heat source; scale bar 50 μm (Supplementary video V1). **b, c)** Top-view schematics of an emulsion droplet formed from a lighter hydrocarbon (pink) and a heavier fluorocarbon (grey) when aligned solely by gravity (**b**) and when a thermal source is passing by (**c**). **d)** Side-view schematic of the same droplet, showing the relevant interfacial tensions γ_H , γ_F , and γ_{FH} . **e)** The droplet's response in a thermal field, which creates an interfacial tension gradient at the droplet-internal capillary surface and results in a steady-state tilt with thermally-induced torque τ_{th} and gravitational torque τ_g being balanced. R_d - droplet radius, R_{cm} - distance between center of rotation and center of mass, F_g - gravitational force.

80 the associated reorientation of the droplets is therefore
 81 primarily driven by the surfactant-free capillary interface
 82 between the two phases. This is readily apparent when
 83 comparing the clearly observable flows in and around
 84 a Janus droplet exposed to a thermal gradient (Fig.2)
 85 with the absence of directional flows around single-phase
 86 droplets made from the emulsion's constituent phases
 87 (Supplementary Fig. S1). We visualize thermally induced
 88 Marangoni flows around the emulsion droplets by dis-
 89 persing fluorescent particles in the surrounding aqueous
 90 medium (Supplementary video V2) and map the flow
 91 fields using particle imaging velocimetry [36]. When sub-
 92 jected to a laser-induced thermal gradient, substantial
 93 fluid motion can be perceived in the vicinity of bi-phase
 94 droplets (Fig. 2a, c). The presence of a small amount of
 95 phase impurities inside some droplets allowed us to qual-
 96 itatively map the flow fields in the fluorocarbon phase

97 (Fig. 2e). The experimentally observed flow patterns are
 98 closely matched by the theoretical flow fields obtained
 99 through finite element modeling (Fig. 2b, d, f), with ex-
 100 perimentally estimated fluid velocities on internal inter-
 101 face and droplet surface assigned as a Dirichlet condi-
 102 tion (Supplementary section II.C). Further, with the ap-
 103 proximated flow field at the droplet interface, we find
 104 that the net force acting on the droplet vanishes, $\vec{F} = \vec{0}$
 105 (Supplementary section II.C) [37]. Single-phase droplets
 106 composed of either emulsion phase exhibit no systematic
 107 external flow (Supplementary Fig. S1), which indicates
 108 that the exterior droplet interfaces do not play a role in
 109 the observed dynamics. The internal interface between

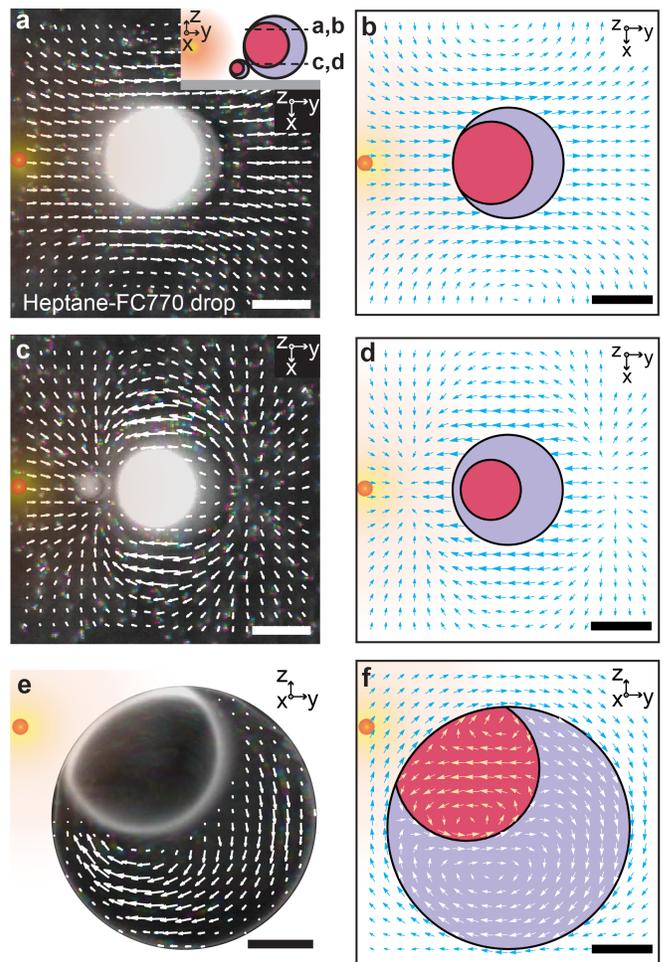


FIG. 2. **Particle imaging velocimetry data and finite element modeling reveal thermocapillary flows around and in bi-phase droplets.** **a, c)** The exterior flows visualized in top view around individual heptane-FC-770 droplets at different height levels match the fields that are obtained by **b, d)** finite element modeling in the same planes. The inset in (a) locates the optical slices shown in (a-d). The location of the IR laser spot is marked schematically with orange dots. **e)** Interior flow fields seen in the FC-770 phase of an 80 μm -sized droplet match **f)** modeled flow fields. All scale bars are 20 μm .

110 the two constituent phases is the critical driver for the
111 manifestation of the observed dynamics.

112 To assess how the curvature of the internal capillary sur-
113 face and the magnitude of the thermal differential affect
114 the droplet dynamics, we systematically varied the dis-
115 tance between the laser spot and droplets with different
116 morphologies and quantified their steady-state tilt angle
117 α^* (Fig. 3). To modify the droplet morphology, we adjust
118 the interfacial tensions between the constituent oils and
119 the aqueous medium by changing the relative surfactant
120 concentrations [5]. Droplets with a large internal inter-
121 face curvature tilt significantly more than similarly-sized
122 droplets with smaller interface curvature (Fig. 3a-c). For
123 all tested droplet configurations, the steady-state rota-
124 tion angle α^* decreased with increasing distance between
125 droplet and heat source (Fig. 3d).

126 For our experiments, we used Fluorinert FC-770 and
127 heptane to form approximately spherical bi-phase emul-
128 sions. FC-770 has a significantly higher density ($\rho_F =$
129 1793 kg m^{-3}) than heptane ($\rho_H = 684 \text{ kg m}^{-3}$), resulting
130 in a pronounced torque, when the droplets' symmetry
131 axis is not aligned with gravity. The rotation of bi-phase
132 droplets in a thermal gradient is opposed by this gravita-
133 tional torque. As the droplets reach the steady-state rota-
134 tion angle α^* the thermally induced torque is balanced
135 by the gravitational torque. The gravitational torque can
136 be mathematically expressed as $\tau_g = R_{\text{cm}} \cdot Mg \sin \alpha^*$,
137 where M is the total mass of the droplet, $g = 9.81 \text{ m s}^{-2}$
138 is the gravitational constant, and R_{cm} is the distance
139 from the droplet's center of rotation to its center of mass
140 (Fig. 1e; details in Supplementary section II.A). This al-
141 lowed us to quantify the magnitude of the thermally in-
142 duced torque as a function of droplet morphology and
143 distance between droplets and heat source (Fig. 3e).

144 The observed dependence of the droplet rotation on the
145 thermal differential and internal interface curvature can
146 be captured in a simple theoretical model: to determine
147 the optically-induced thermal torque and steady-state rota-
148 tion angle α^* for a given droplet geometry, we minimize
149 the sum of gravitational energy $E_g(\alpha)$ and the energy
150 $E_{\text{th}}(\alpha)$ resulting from the thermally induced variation in
151 interfacial tension across the droplet-internal capillary in-
152 terface with respect to the droplet's tilt angle α (detailed
153 discussion in Supplementary section II.B) [38–40]. This
154 approach is equivalent to balancing gravitational torque
155 $\tau_g(\alpha)$ and thermally induced torque $\tau_{\text{th}}(\alpha)$. The two en-
156 ergies can be expressed as

$$E_g(\alpha) = R_{\text{cm}} Mg (1 - \cos \alpha) \quad (1)$$

$$E_{\text{th}}(\alpha) = \int_{\Omega_i} \gamma_{\text{FH}}(T(\vec{r}, \alpha)) dA. \quad (2)$$

157 The shape of the curved capillary interface Ω_i between
158 the droplet's constituent phases can be described as a
159 spherical cap with surface area $A_i = 2\pi R_i^2 (1 - \cos \theta_i)$,
160 where R_i is the radius of curvature of the internal in-
161 terface and θ_i represents the spherical cap's apex half
162 angle (Fig. S4). To allow for an analytical solution, we
163 linearize the surface tension dependence on temperature

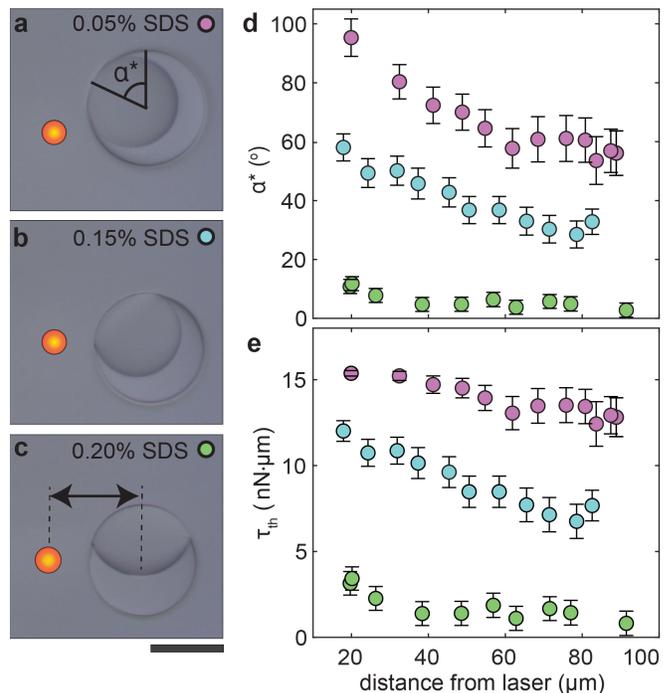


FIG. 3. **Quantifying rotation in thermal gradients for different droplet morphologies.** a-c) Side view images of droplets in the vicinity of a focused NIR laser in 0.4 wt% Zonyl and varying sodium dodecyl sulfate (SDS) concentrations to vary interface curvature - 0.05 wt% SDS (a, magenta), 0.15 wt% SDS (b, cyan), 0.20 wt% SDS (c, green); scale bar 50 μm . d) Steady-state rotation angle α^* of droplets in (a-c) and e) the corresponding thermal torque τ_{th} vs. distance from the laser spot. Morphological droplet parameters are listed in the Supplementary (table S2).

164 $\gamma_{\text{FH}}(T) = \gamma_0 + \gamma_1 T$ and approximate the spatial tem-
165 perature profile in a co-rotating droplet reference frame
166 as $T(\vec{r}, \alpha) = T_0 + \vec{r} \cdot \vec{n}(\alpha) \frac{\Delta T}{2R_d}$. Here, R_d is the droplet
167 radius, \vec{r} the position vector measured from the droplet
168 center, $\vec{n}(\alpha)$ the direction of the thermal gradient in the
169 co-rotating reference frame, and ΔT the temperature dif-
170 ferential across the droplet. The steady state rotation
171 angle α^* can then be expressed analytically as

$$\tan \alpha^* = \frac{\gamma_1 \frac{\Delta T}{2R_d} A_i (d - \frac{1}{2} R_i (1 + \cos \theta_i))}{R_{\text{cm}} Mg}, \quad (3)$$

172 where d is a morphological parameter that describes the
173 distance between the droplet center and the center of in-
174 terface curvature (derivations in Sup-
175 plementary sections II.A, B).

176 By applying this simple model to the experimentally
177 studied droplet morphologies shown in Fig. 3, we recover
178 the observed behavior qualitatively. The droplets all ex-
179 hibit a very similar gravitational energy dependence on
180 the rotation angle α (Fig. 4a), since their mass and center
181 of gravity are very similar. However, they show an order
182 of magnitude difference in their surface energies due
183 to their different capillary interface curvatures (Fig. 4b).

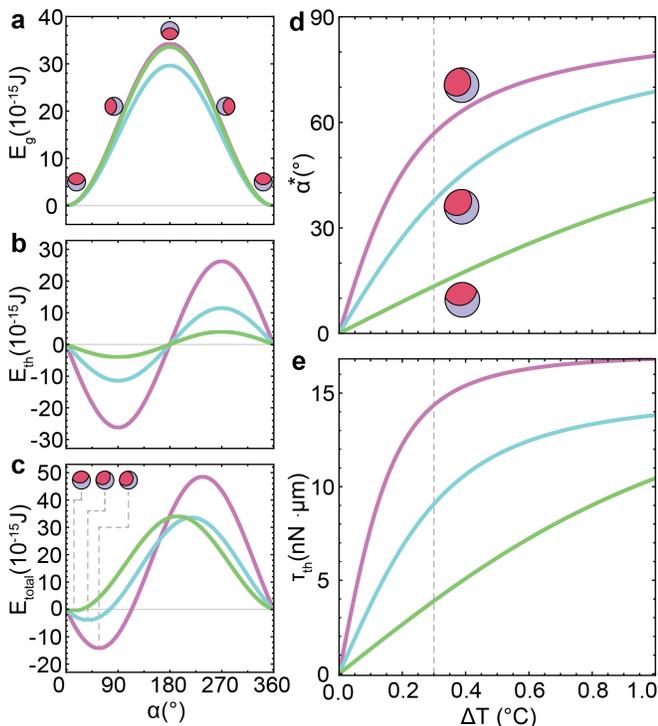


FIG. 4. **Modeling of droplet response: energy balance, resulting rotation angles, and torques.** a-c) Potential energy E_g due to gravity, energy E_γ due to thermally induced surface tension gradient, and the sum of both vs. rotation angle α . d, e) Steady state rotation angle α^* and thermal torque τ_{th} vs. temperature differential ΔT across droplet. The three differently shaded curves represent droplet geometries with increasing internal interface curvature that match the experimentally observed geometries shown in Fig. 3; $R_i = 0.82R_d$ (magenta), $R_i = 0.86R_d$ (cyan), $R_i = 0.99R_d$ (green), $\gamma_1 = -0.2 \text{ mN m}^{-1} \text{ K}^{-1}$.

This shifts the global minimum in the rotational energy landscape away from 0° towards larger tilts (Fig. 4c). In agreement with experiments (where the temperature differential decreases with distance from the laser spot), the orientation angle grows monotonically with increasing temperature differentials for all shown morphologies until it saturates at 90° (Fig. 4d). Correspondingly, the thermal torque grows as well (Fig. 4e).

The droplets' dynamic response to thermal gradients is observable at a laser power of $\sim 50 \text{ mW}$ without any absorption-enhancing dye in the aqueous medium. In addition, we observe small translational motion, if droplets are exposed to significantly higher laser power ($\sim 150 \text{ mW}$, see video V3 and Fig. S3), which suggests that the thermal differentials required to induce droplet rotation are significantly smaller than the temperature gradients needed for single-phase droplet manipulation [27, 28].

To estimate the magnitude of the thermal gradient needed to induce droplet reorientation, we harnessed the temperature-dependent fluorescence of 2',7'-

dichlorofluorescein (FL27) [41, 42]. Unlike other common organic dyes, whose fluorescence decreases with increasing temperature [43], FL27 exhibits increased light emission in the Anti-Stokes band for higher temperatures (when excited with green light of 532 nm wavelength). This positive correlation of temperature and Anti-Stokes emission can clearly be seen (Fig. 5a), when collecting the emitted light with a short-pass filter (cutoff at 520 nm). The short-pass filter suppresses elastically scattered light and Stokes band fluorescence, which permits optical quantification of small thermal gradients.

Knowing the dyes' emission strength as a function of temperature, we mapped the thermal field around the near-IR laser focus (Fig. 5b). A NIR-absorbing dye was added to the aqueous medium in these experiments in order to increase the thermal gradient, so that it could be visualized with this optical technique. Therefore, the gradient measured in this experiment is significantly larger than in the previous experiment and could be used to rotate a larger droplet. Although the raw intensity data is affected by substantial pixel noise, radial averages around the laser's focal center provide a reasonable estimate of the temperature of the aqueous medium as a function of distance from the laser focus (Fig. 5c), which is well approximated with a simple power law (Supplementary section I.D). This allowed us to quantify the thermal differential that the droplets were experiencing as a function of distance to the laser for various values of laser currents. With this knowledge, we quantify the correlation between the temperature differential ΔT across a droplet and its steady state tilt (Fig. 5d-f), which we determined by side-view optical microscopy. The measured steady state rotation angle α^* is used to quantify the gravitational torque, which is equal in magnitude to the thermal torque (Fig. 5g). Theoretical predictions from our simple analytical model obtained by substituting the measured temperature profile in the equation of the thermal energy (eq. (2)) are in good quantitative agreement with the experimental observations for rotation angle and torque (Fig. 5f, g). The thermal differential needed to induce droplet rotation is smaller than 0.05°C across the diameter of the droplet.

In summary, Janus emulsion droplets dynamically reorient in a thermal gradient. These rotations are due to thermocapillary effects occurring at the droplets' internal interface. A temperature gradient across the surfactant-free internal interface generates an interfacial tension differential that induces Marangoni-type fluid motion within the droplets, entraining flows around them. With the internal interface being the driver for the droplet reorientation, droplet morphology strongly influences the response to thermal gradients: Droplets with highly curved interfaces rotate more than those with a flatter internal capillary surface. Bi-phase emulsion droplets with a surfactant-free internal interface thus harbor a built-in motor that is activated via a temperature differential across the droplet. The rotational effect of thermocapillary motion in surfactant-stabilized multi-phase droplets

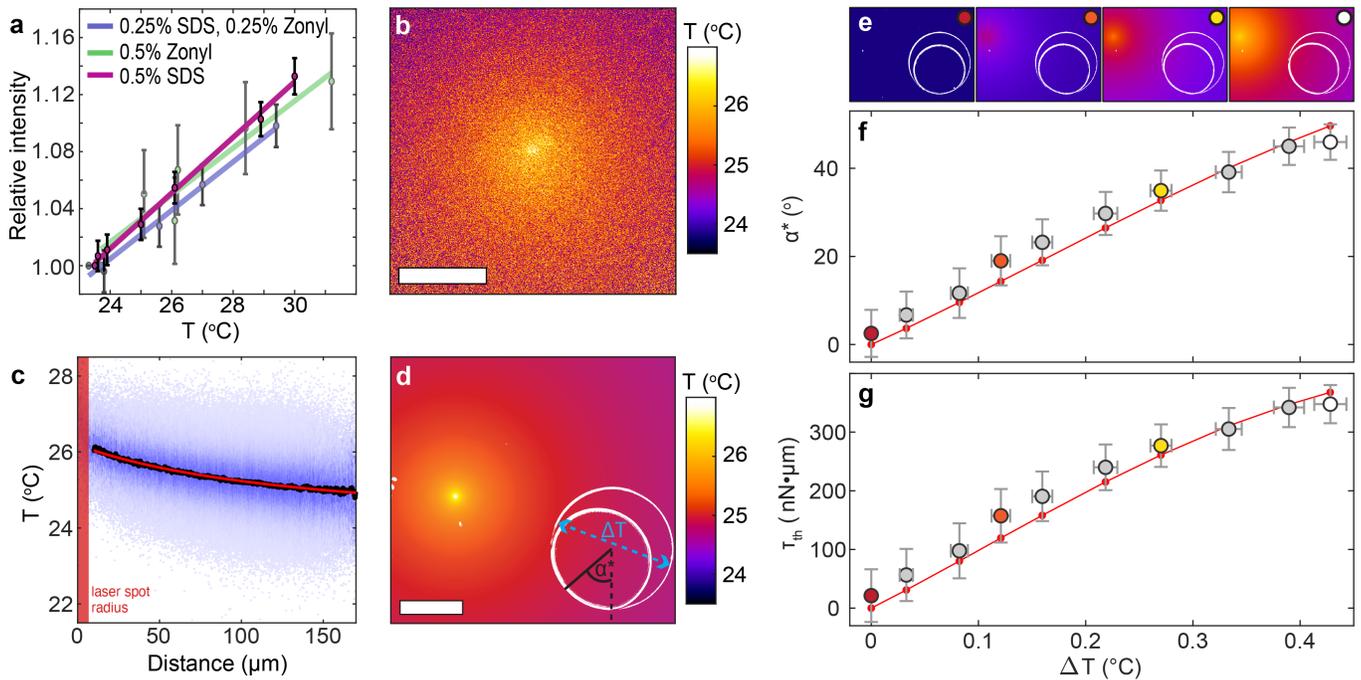


FIG. 5. **Quantifying droplet rotation and torque as a function of temperature differential.** **a)** Temperature dependence of emission intensity of Fluorescein 27 with different SDS and Zonyl surfactant concentrations in the aqueous medium relative to emission at room temperature. **b)** Temperature distribution around laser measured from the relative intensity of Fluorescein 27 using the fit shown in (a); scale bar: 100 μm . **c)** Measured temperature (blue) as a function of distance from the laser with mean values (black) and fit (red). **d)** Temperature map around laser spot using the fit shown in (c) with optical micrograph overlay of droplet; scale bar 100 μm . **e)** Optical micrograph overlays of fitted temperature distribution for laser currents of 0.5 A, 0.8 A, 1.1 A and 1.4 A. **f)** Rotation of droplet as a function of temperature differential ΔT across the droplet, as shown in (d). **g)** thermal torque τ_{th} vs. ΔT . Data points shown in color in (f, g) correspond to images in (e) marked with the same color. Experimental observations match the theoretically predicted behavior (red curves).

described here for the first time could prove important in micro-fluidic sorting, droplet mixing, micro-reactors, thermally controlled adaptive micro-optical systems, and opto-fluidic calorimetric sensors that translate micron-scale heat flows into optically detectable deflections of light.

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