ABSTRACT: Driven by nanoparticle plasmonic heating, sessile droplet evaporation presents challenges on the coupling mechanisms between time-spatial heat source distribution and flow/temperature fields in a droplet. Here, symmetric/asymmetric solar-driven droplet evaporation is investigated. An infrared camera captures droplet surface temperatures in the micrometer scale after correction. An optical three-dimensional profiler quantifies nanoparticle deposition in the nanoscale. We show that droplet surface temperatures do display a nonmonotonic variation trend. Based on measurements, we are able to decouple the droplet into a contact line region (CLR) and a bulk volume region (BVR). The CLR volume is two to three magnitudes smaller than the droplet volume. The temperature gradient is significant in CLR, but flat temperature exists in BVR. Radial flow in BVR transports nanoparticles from the droplet body to the contact line, while Marangoni flow in CLR stabilizes nanoparticles there. Light energy is also decoupled based on its wavelength band. It is found that CLR dominates the visible energy absorption, but BVR has a weak contribution. Top light heating causes symmetry temperatures and a coffee-ring profile along the circumference. However, side heating yields higher temperatures and more nanoparticles deposition on the sunny side than on the night side. The above findings are valid when the initial droplet volume and incident irradiation flux are changed on the hydrophilic wall and hydrophobic wall maintain the evaporation modes of constant contact flow/temperature fields in the droplet. The hydrophilic wall and hydrophobic wall maintain the evaporation modes of constant contact diameter and “stick—slip”, respectively. The present paper enhances the understanding of light-induced droplet evaporation from the multiscale point of view.

INTRODUCTION

Interaction of light with a metal involves both the surface plasmon polariton (SPP) occurring on the metal surface and localized surface plasmon resonance (LSPr) confined in a local space around isolated nanoparticles. One of the LSPr effects is to generate heat in the nanoscale, driving much attention due to various applications in chemical reactions, cancer treatment, and solar energy extraction. In this paper, light-driven nanofluid droplet evaporation is investigated, which is seldom reported previously. A xenon lamp emits light covering a wide wavelength band range to simulate nature sun. Light energy would be absorbed by nanoparticles in the droplet, which alters the flow/temperature fields in the droplet. Such a problem becomes more complicated compared with substrate heating-induced droplet evaporation.

Here, we start from a short description of substrate heating-induced droplet evaporation. Then, we describe the coupling between plasmonic heating and flow/temperature fields in the droplet, which helps us understand the phenomena and the underlying mechanism. Previously, abundant studies have been performed on substrate heating-induced droplet evaporation. For a sessile droplet, droplet temperatures decrease from the bottom to the apex. Radial flow and Marangoni flow are established by nonuniform evaporation fluxes on the droplet surface and temperature gradient in the droplet, respectively. They take place in the droplet length scale, which are coupling with each other. When nanofluid is used, the flow-induced particle deposition forms various patterns such as a coffee ring on the substrate. Thokchom et al. have shown that, for asymmetric substrate heating, the temperature of the free surface decreases from the heating side to the nonheating side. The flow field in the droplet results in particle deposition in the colder region of the substrate. Generally, the substrate heating-induced droplet evaporation involves weak or no coupling between the heat source and flow/temperature fields. Such a problem is still not well solved, presenting several challenges to be addressed.

Several articles deal with infrared (IR) radiation-driven droplet evaporation, in which only Thokchom et al. discuss the flow/temperature fields in the droplet. IR can be
absorbed by the water surface, acting as a thin liquid film heat source. For IR falling on a droplet surface, temperatures are higher at the apex and lower at the contact line. Such a temperature gradient generates Marangoni flow in the droplet length scale. The flow direction is inverse to that caused by substrate heating. In their work, micrometer-sized particles act as the tracer for flow visualization, which have a weak influence on IR absorption.12

Solar light includes not only IR energy but also visible energy. When solar light irradiates a sessile nanofluid droplet, IR energy is absorbed in the base liquid, and visible energy is absorbed by nanoparticles due to the LSPR effect. The problem presents complicated coupling between nanoparticle plasmonic heating and flow/temperature fields in the droplet. Assuming a uniform nanoparticle distribution in the droplet initially, light irradiation will establish internal flows, reorganizing nanoparticles to influence plasmonic heating intensity. The nanoparticle redistribution is influenced not only by bulk flow in the droplet but also by various microscopic disturbances. Many interaction forces dominate the microscopic motion of nanoparticles.15,16 Van der Waals forces arise between all molecules and particles.16 Magnetic/electrostatic force, repulsive force, confining or jamming forces, solvation force, structural force, and capillary force are other examples that influence the motion of particles.15 The Brownian movement of particles is caused by all the impulses of fluid molecules on the particle surface.17 The above forces redistribute nanoparticles, alter the distance between nanoparticles, and even result in the coalescence of nanoparticles to form a particle cluster. Because the plasmonic heating by individual nanoparticle is dependent on the gap between nanoparticles,18 the nanoparticle reassembly dominates the heat source distribution in different length scales. In turn, the plasmonic heat generation influences the flow/temperature fields in the droplet.

Recently, droplet evaporation is experimentally studied with solar light falling on the droplet surface, focusing on the analysis of critical nanofluid concentration.19 With the increase in initial nanofluid concentration \( c_0 \), droplet evaporation rates display a rising regime and a constant regime. The two regimes are interfaced at a critical \( c_c = 10 \text{ ppm} \), corresponding to sufficient nanoparticle deposition for visible energy absorption near the contact line. A larger \( c_c \) than the critical value results in redundant nanoparticle deposition beneath effective particle layers. Only the effective particles contribute to plasmonic heating, but the extra nanoparticles do not, explaining the critical phenomenon.

Here, we investigate the solar-driven nanofluid droplet evaporation, which widely occurs in nature and engineering such as water-steam generation and solar collector.20 Compared with substrate heating or IR heating, the studied problem in this paper is complicated and not understood due to strong coupling between nanoparticle plasmonic heating and flow/temperature fields. Each nanoparticle can be considered as a point heat source in the nanoscale. All the point heat sources are dynamically moving. Thus, the problem displays strong multiscale characteristic. To explore the phenomena and mechanisms in the multiscale framework, the paper is organized as follows. Experiment Setup and Methods describes the experimental setup, including droplet surface temperature measurement. Such measurement in the micrometer scale near the contact line forms the basis to decouple the droplet into different length scales. Results and Discussion contains four sections. The first section discusses droplet evaporation dynamics, in which the dynamic \( V-t \) curves are useful for energy analysis in the latter section, where \( V \) and \( t \) are the droplet volume and time, respectively. The second section reports the decoupling of length scales, corresponding to different flow/temperature behaviors. The third section presents mechanism analysis from the multiscale point of view. The fourth section deals with the effect of various parameters on droplet evaporation. The major conclusions are summarized in Conclusions.

**Experiment Setup and Methods**

**Experimental Setup.** Two arrangements of the experiment setup are shown in Figure 1a for top light heating and Figure 1b for side light heating. Both arrangements include a high-speed camera (1; VW-9000, Keyence), a xenon lamp as the solar simulator (2; CEL-SS00), a PMMA glass cloak (3), a microsyringe pump (4; RSP01-BDG), a high-speed IR camera (5; ImageIR S380, InfraTec), and a glass substrate unit (6 and 7) to support a droplet (8). For top light heating, the high-speed camera and IR camera are horizontally positioned with a relative angle of 90°. Alternatively, for side light heating, the IR camera vertically faces the droplet surface. Both the light source and high-speed camera face the droplet from the side direction, with a relative angle of 90°. The droplet and its supporting unit are centrally located in the cloak, protecting the droplet from air disturbance. Two holes are arranged in the cloak, ensuring the light source and optical instruments directly facing the droplet.

Nanofluid ejection is controlled by the microsyringe pump to form a hanging droplet under the syringe needle. A sessile droplet is formed after having the droplet contact the glass substrate and receding the syringe needle from it. The initial nanofluid droplet volume \( V_i \) is in the range of 1–3.0 \( \mu \text{L} \). The xenon lamp has a similar spectrum distribution as solar light. An AM 1.5 filter is used to eliminate stray light, making the spectrum of incident light further approach that of
nature sunlight (see Figure S1 of the Supporting Information). The radiation intensity \( q \) on the droplet can be varied in a wide range. Three \( q \) values are selected as 0 (nature light in the laboratory environment), 800, and 1400 W/m\(^2\) in this paper. The diameter of the light spot is \( \sim 80 \text{ mm} \), which is at least one magnitude larger than the droplet contact diameter \( D = 1.5 - 2.5 \text{ mm} \). The droplet is located at the light spot center. For both top heating and side heating, the distance between the light filter and droplet is kept as \( \sim 85 \text{ mm} \). The high-speed camera with a 4.7 \( \mu \text{m} \) spatial resolution captures the droplet morphology. Contact diameter \( D \) and contact angle \( \theta \) are obtained after processing image files. \( D \) has an uncertainty of 4.7 \( \mu \text{m} \), equivalent to the high-speed camera resolution. To evaluate the \( \theta \) uncertainty, several sessile droplets are statically measured by both a high-speed camera and a contact angle meter (Dataphysics OCA20), whose uncertainty is 0.1°. The maximum deviation between the two measurements is less than 0.4°. Thus, the uncertainty of \( \theta \) is 0.5°. In this study, the parameters of the droplet morphology are subject to measurement. The IR camera captures the droplet surface temperature in a 3–5 \( \mu \text{m} \) band range, having a spatial resolution of 24 \( \mu \text{m} \) and a temperature resolution of 0.02 K. The temperature correction near the contact line described in Measurement of Droplet Surface Temperature does not change the spatial resolution of temperature measurement. The high-speed camera and IR camera are synchronized to function in a time resolution of 20 ns. The image recording rate is 20 frames/s, which is sufficiently fast for slow transition of droplet dynamics in this study.

Droplet evaporation induces coffee-ring deposition on the glass substrate. For side heating, coffee-ring parameters are nonuniform along the circumference. The width \( W \) and height \( h \) of the coffee ring are measured by an optical 3D profiler (Bruker Contour GT-K Elite) to have resolutions of 75 \( \text{nm} \) in the width direction and 0.2 \( \text{nm} \) in the height direction. Resolutions are at least two magnitudes smaller than practical values, ensuring precise measurements. During the experiment, the environment temperature and air humidity are controlled to \( T_e = 25 \pm 1 \text{ °C} \) and \( q = 30 \pm 2 \% \), respectively. Uncertainties of various parameters are available in Table S1 of the Supporting Information.

The Nanofluid. Base materials used to prepare nanofluid are gold chloride hydrate (HAuCl\(_4\) Au ≥ 49%) and trisodium citrate (Na\(_3\)C\(_6\)H\(_5\)O\(_7\) C and \( \theta \)). Provided by Sigma-Aldrich (USA). Gold nanofluid is formed by mixing and reacting the two materials in boiling water.24 Nanofluid stabilization is enhanced by ultrasonic oscillation. The detailed characterizations of nanoparticles are given in Figure S2 of the Supporting Information, including nanoparticle image, size distribution, zeta potential, and transmittance. Nanoparticles display quasi-normal distribution with an average diameter \( d_p = 18.87 \text{ nm} \) by analyzing 138 particles. The zeta potential is \(-67.5 \text{ mV} \), which has excellent stabilization.25 Transmittance \( \tau_e \) characterizes nanofluid optical performance, reaching the minimum at \( \lambda = 520 \text{ nm} \). The \( \tau_e \) values decrease with the increase in \( \varepsilon \) from 10 to 50 ppm.

Measurement of Droplet Surface Temperature. Challenge exists for droplet surface temperature measurement including near the contact line in micrometer resolution. Only having such fine measurement, we are able to decouple the whole droplet into different length scales. This section presents a correction method based on IR temperature measurement. The IR camera measures temperatures based on received infrared energy in the 3–5 \( \mu \text{m} \) wavelength from an object. For top light heating, the IR camera measures droplet surface temperature \( T_{IR} \) directly from the glass substrate \((D > \lambda/4)\). Referring to the second part from the contact line (CL) to point C, the liquid penetration length can be corrected to reach 0.99 at \( L_{IR} = 300 \text{ μm} \). The maximum deviation between the two measurements is \( T_{IR} \). Based on Lambert–Beer law,24 Figure 2a plots \( I/I_0 \) versus \( L_0 \), which decreases significantly with the increase in \( L_0 \) to reach 0.01 at \( L_0 = 300 \text{ μm} \) (see the determination of \( I/I_0 \) and Figure S3 in the Supporting Information). The IR camera captures the water surface temperature in a 300 μm-thin liquid film but cannot detect emission signals beyond 300 μm. Thus, \( L_{IR} = 300 \text{ μm} \) is defined as the critical measurement length. Based on Kirchhoff’s law,26 emissivity \( \varepsilon \) increases with the increase in \( L_0 \) to reach 0.99 at \( L_{IR} = 300 \text{ μm} \).

![Figure 2](https://dx.doi.org/10.1021/acs.langmuir.9b03122Langmuir.2020,36,1680-1690)
where $\sigma$ is the Stefan–Boltzmann constant: $\sigma = 5.67 \times 10^{-8} \, \text{W/(m}^2\cdot\text{K}^4)$. $E_i$ is

$$E_i = \sigma \varepsilon_i (T_i + 273.15)^4 e^{-\beta L}$$

(3)

where $\varepsilon_i$ is the glass substrate emissivity, $T_i$ is the glass substrate temperature, and $\beta$ is the average absorption coefficient of liquid in the 3–5 $\mu$m wavelength ($\beta = 15.2 \, \text{mm}^{-1}$).$^{23}$ $E_i$ is

$$E_i = (1 - e^{-\beta L})\sigma (T_i + 273.15)^4$$

(4)

Combining eqs 1–4 yields

$$T_i = \frac{(T_m + 273.15)^4 - \varepsilon_i (T_i + 273.15)^4 e^{-\beta L}}{(1 - e^{-\beta L})} - 273.15$$

(5)

Equation 5 establishes the connection between $T_i$ and $T_m$. $T_i$ is the substrate temperature measured by a thermocouple, which is ~3 mm away from the nearest contact line of the sessile droplet. $T_m$ represents the actual temperature near the contact line due to uniform temperature distribution from the contact line to the thermocouple.

To determine $\varepsilon_i$, the substrate temperature is also measured by the IR camera, which is recorded as $T_{i,IR}$. Due to the non-black-body of the substrate, $T_{i,IR} \neq T_i$. Thus, $\varepsilon_i$ is written as$^{24}$

$$\varepsilon_i = \left(\frac{T_{i,IR} + 273.15}{T_i + 273.15}\right)^4$$

(6)

The measurement of $T_{i,IR} = 12.6 \, ^\circ\text{C}$ and $T_i = 25.2 \, ^\circ\text{C}$ yields $\varepsilon_i \approx 0.84$. $L_i$ in eq 5 is determined by the droplet image file, which is recorded by a VH-SOL lens. The droplet morphology is represented by a curve fitting equation, yielding an uncertainty of 4.7 $\mu$m for $L_i$, which is equivalent to the spatial resolution of the lens (see Figure S4 of the Supporting Information). Such uncertainty of $L_i$ is included in later analysis.

It is noted that eq 5 can determine the corrected temperature excluding CL due to infinite $T_i$ with $L_i \approx 0$ at CL. Figure 2d shows a strategy to solve this issue. The point A on the droplet surface is specified, whose radial distance from CL equals to one spatial resolution of 24 $\mu$m of the IR image file. Substituting $L_{i,A} \neq 0$ into eq 5 obtains $T_{i,A}$. The corrected temperature at CL ($T_{CL}$) is achieved by extrapolation based on $T_{i,A}$. It is observed that $T_m$ and $T_i$ coincide with each other in the major center region (first part), but a large difference between them exists near the CL region (second part). The deviation reaches the maximum at CL having $T_i = 32.1 \, ^\circ\text{C}$ and $T_m = 24.9 \, ^\circ\text{C}$. Thus, we conclude that the correction technique is necessary for shorter liquid film thickness than the critical value.

## RESULTS AND DISCUSSION

### Light-Driven Droplet Evaporation Dynamics

Contact diameter $D$ and contact angle $\theta$ characterize droplet evaporation dynamics. Droplet volume $V$ at time $t$ is

$$V = \frac{\cos^3 \theta - 3 \cos \theta + 2}{24 \sin^3 \theta} \pi D^3$$

(7)

To operate the system, the syringe pump controls the initial droplet volume $V_i$ by setting a specific value such as 1, 2, or 3 $\mu$L. Because the setting value contains a larger uncertainty, eq 7 is used to achieve accurate $V_i$ based on measurements of $D_i$ and $\theta_i$. Having uncertainties of 4.7 $\mu$m for $D_i$ and 0.5$^\circ$ for $\theta_i$, we obtain the uncertainty based on the error transfer theory, which is 0.016 $\mu$L for $V_i \approx 1 \, \mu$L, 0.030 $\mu$L for $V_i \approx 2 \, \mu$L, and 0.034 $\mu$L for $V_i \approx 3 \, \mu$L. These uncertainties yield very small relative errors of 1.6, 1.5, and 1.1% for the three setting values of 1, 2, and 3 $\mu$L, respectively. Thus, $V_i$ values determined by eq 7, instead of the setting value, are marked in the figures. It is noted that such a relative error increases with time evolution due to the increased relative error of contact angle $\theta$, but the maximum relative error of $V$ does not exceed 11.3% when $\theta$ reaches 5$^\circ$.

The experimentally determined $V$–$t$ curves are used for multiscale energy analysis in Multiscale Mechanisms for Symmetry/Asymmetry Droplet Evaporation. Both top heating and side heating produce similar droplet dynamics. During evaporation, for top heating, the projected area perpendicular to the light irradiation direction is constant, but for side heating, it decreases due to the decreased droplet height. The droplet lifetime is ~400 s for top heating and ~600 s for side heating, indicating faster evaporation for top heating than side heating (see Figure 3).

![Droplet evaporation dynamics](https://dx.doi.org/10.1021/acs.langmuir.9b03122)

Figure 3. Droplet evaporation dynamics at $c_i = 10$ and 50 ppm ((a) for top light heating and (b) for side light heating).

A previous study focuses on substrate heating-induced droplet evaporation, having two modes of constant contact diameter (CCD) and constant contact angle (CCA).$^{26}$ CCD occurs at larger contact angle hysteresis to pin the contact line,$^{27}$ but CCA occurs for weak wall–liquid interaction so that the contact line recedes from its initial location.$^{28}$ Here, the static contact angle of the droplet on the substrate is ~80$^\circ$, but the contact angle hysteresis attains ~40$^\circ$. For light-driven droplet evaporation, two mechanisms ensure the contact line pinning to achieve CCD mode: larger contact angle hysteresis on hydrophilic wall and enlarged wall roughness due to nanoparticle deposition. Droplet evaporation on the hydrophobic wall is also studied in this paper.

Droplet lifetime consists of two stages. The first stage sustains the period of 0 to $\tau_c$. The droplet height decreases to form a thin liquid film at $t = \tau_c$. Due to insufficient pinning energy barriers at the contact line to balance residual surface energy of the liquid film, the liquid film suddenly breaks up and recedes from the initial contact line. This process reorganizes the residual liquid to form a droplet having a smaller diameter.
than the initial one. Light continues to irradiate the droplet until it dries out. Thus, two nanoparticle deposition patterns are observed: a coffee ring corresponding to the first stage and a nanoparticle island corresponding to the second stage.

For both light heating modes, almost identical droplet dynamics are observed for \( c_i = 10 \) and 50 ppm (see Figure 3). Our recent work on top heating explains the insensitive effect of nanofluid concentrations on droplet evaporation rates for \( c_i > 10 \) ppm. With continuous increase in \( c_i \) from pure water to a larger value such as 100 ppm, droplet evaporation rates increase first until a critical condition is reached, beyond which evaporation rates are unchanged. This variation trend is related to nanoparticle plasmonic heating of a coffee ring. The critical concentration \( c_{i,c} = 10 \) ppm corresponds to sufficient nanoparticle deposition. Further increase in \( c_i \) generates extra nanoparticles beneath the effective nanoparticle layers to have no contribution to heat generation, which is called the shadow effect. Here, the critical phenomenon is also observed for side heating. Because we focus on symmetry/asymmetry features of light-induced droplet evaporation, only two \( c_i \) values are used. The \( V-t \) curves can be found in Figure S5 of the Supporting Information.

**Symmetric/Asymmetric Droplet Surface Temperatures.** For side heating, it is not surprising to observe higher temperatures on the sunny side than on the night side. However, for the first time, we experimentally find the nonmonotonically varied droplet surface temperatures, which are thoroughly different from the monotonic variation trend for substrate heating or IR irradiation conditions. Based on our measurements, we could decouple the whole droplet into two length scales including the contact line region (CLR) and bulk volume region (BVR). The former is two to three magnitudes smaller than the latter. The significant temperature gradient in CLR and flat temperature distribution in BVR are newly recognized as a boundary layer effect for light-driven droplet evaporation. The heterogeneous coffee ring is also reported.

Figure 4 illustrates droplet surface temperatures for both heating modes, noting that temperatures in Figure 4b for side heating are corrected based on eq 5. Generally, top heating creates higher temperatures than side heating. Side heating has higher temperatures near the contact line on the sunny side than on the night side (see the image at \( t = 450 \) s in Figure 4b). These temperatures are replotted in Figure 5. Top heating is treated first (see Figure S5a,b). An original point \( O \) is defined at the droplet apex, and \( L_c \) is the arc length along the droplet surface starting from point \( O \). Two regions are observed: a higher temperature region within \( \delta \) and a quasi-flat temperature region beyond \( \delta \). The determination of \( \delta \) is similar to the thermal boundary layer thickness described in heat transfer textbook:

\[
\frac{T_{CL} - T_{\delta}}{T_{CL} - T_O} = 95\%
\]

(8)

where \( T_{CL} \) is the temperature at the contact line, \( T_{\delta} \) is the temperature at the crossing point adjoining the two regions, and \( T_O \) is the temperature at the droplet apex (\( L_c = 0 \)).

The determination of \( \delta \) for side heating (see Figure 5c,d) is similar to top heating. The only difference is the coordinate used. For side heating, the original point \( O \) is at the droplet apex, but the radial coordinate is used as the horizontal coordinate. To explore asymmetry characteristics of temperature, \( \delta_1 \) and \( \delta_2 \) are defined for the left side and right side, respectively. For top heating, the two values are almost identical to display symmetry behavior. For side heating, \( \delta_1 \) and \( \delta_2 \) refer to the sunny side and night side, respectively. The sunny side creates larger \( \delta \) and higher temperatures within \( \delta \) compared with the night side, displaying asymmetry characteristic. The recognition of the two temperature regions indicates the boundary layer effect, which is distinct for light-driven
nanofluid droplet evaporation but does not exist in substrate heating cases.\textsuperscript{10}

The parameters \((\delta/L_{\mathrm{CL}})^3\) for top heating and \((\delta/R)^3\) for side heating represent the ratio of CLR volume with respect to the whole droplet volume, where \(L_{\mathrm{CL}}\) is the arc length along the droplet surface starting from point \(O\) to the contact line, and \(R\) is the contact radius \((R = 0.5D)\). Physically, \((\delta/L_{\mathrm{CL}})^3\) and \((\delta/R)^3\) scale the thermally influenced volume near the contact line. The whole droplet is decoupled into two regions of CLR and BVR. The former corresponds to a smaller length scale \(\delta\) with a significant temperature gradient, and the latter refers to a larger length scale with flat surface temperature. For top heating, CLR contributes 0.6–3\% of the total droplet volume in lifetime. The CLR volume relative to the droplet decreases versus time, and the curves in Figure 5b almost coincide with each other for the left side and right side due to symmetry behavior. Generally, \((\delta/L_{\mathrm{CL}})^3\) decreases versus time at first, but approaches a constant value for \(t > 180\) s, indicating the unchanged thermally influenced volume in the latter stage. An exception is observed to have an increase for \(c_i = 50\) ppm at \(t = 300\) s. This is due to the additional heat generation by nanoparticles populated elsewhere beyond the coffee ring when more nanoparticles are involved (see Figure 6b).

Comparatively, for side heating, CLR contributes 0.1–1.4\% of the droplet volume in lifetime. The sunny side has a significantly larger CLR volume than the night side. Besides, \((\delta/R)^3\) shows an apparent increasing trend with respect to time, especially for the sunny side.

Figure 6 explores circumferential angle-dependent coffee-ring profiles, in which four locations are marked as 1–4, noting that 4 in Figure 6c,g corresponds to the sunny side. Usually, these points are selected to face the center of sunny/night side. In case there is a particle island there, the selected point should deviate from the particle island. Thus, it may not be at the center of sunny/night side (see Figure 6c). The triangular/trapezoidal cross section can be characterized by width \(W\) and height \(h\). \(W\) is in the range of 3–4 \(\mu\)m, but \(h\) is different for different cases. Top heating yields quasi-uniform circumference ring profiles \((h_1 \approx h_2 \approx h_3 \approx h_4)\), but side light heating creates a higher cross-section profile on the sunny side than on the night side (largest \(h_4\) among \(h_1 \sim h_3\)), showing asymmetric nanoparticle deposition. For identical droplet volume at \(t = 0\), the droplet with \(c_i = 50\) ppm contains \(\sim 5\) times of nanoparticles than that with \(c_i = 10\) ppm. Because most of nanoparticles should deposit on the coffee ring, the \(c_i = 50\) ppm case results in a much higher cross-section profile than the \(c_i = 10\) ppm case (see Figure 6).

Multiscale Mechanisms for Symmetry/Asymmetry Droplet Evaporation. This section presents theoretical analysis to explain the multiscale feature and the distinct coffee-ring structure for symmetric/asymmetric light-driven droplet evaporation.

Multiscale Flow in the Droplet To Induce Nanoparticle Deposition. Two types of flows are involved in the droplet: radial flow in the droplet length scale and Marangoni flow in the short length scale of CLR (see Figure 7). Radial flow is established by nonuniform evaporation flux \(J\) on the droplet surface, which is larger near the contact line than elsewhere.\textsuperscript{7} Radial flow entrains nanoparticles to flow from the droplet

\[
\frac{\delta}{\delta R} \leq 0.5
\]

Figure 7. Multiscale light-driven droplet evaporation: (a) physical model of visible energy absorption for top light heating; (b) physical model of visible energy absorption for side light heating; (c) flow and energy analysis for top light heating; (d) flow and energy analysis for side light heating. The total energy is decoupled into infrared energy absorption in a thin-film region I and visible energy absorption in regions I and II. Marangoni flow and radial flow occur in CLR and BVR, respectively. Extra particles are hidden in the vertical direction for top heating, and extra particles are hidden in the radial coordinate for side heating.

Figure 6. Coffee-ring profile parameters (a), (b), (e), and (f) for top light heating showing quasi-uniform distribution at different ring locations; (c), (d), (g), and (h) for side light heating showing stronger deposition on the sunny side than on the night side. The \(x\) and \(z\) axes represent horizontal and vertical directions, respectively.)
center to the contact line, which is key for nanoparticle deposition in CLR. Radial flow velocity \( u_r \) is given by

\[
u_r = \frac{J}{\rho_l} = \frac{D_k (c(T) - c_\infty)}{R \rho_l} (1 - \frac{r}{R})^{-5}
\]

where \( \rho_l \) is the liquid density, \( D_k \) is the vapor diffusion coefficient \( (D_k = 2.54 \times 10^{-5} \text{ m}^2/\text{s}) \), and \( c(T) \) is the saturation vapor concentration at droplet surface temperature \( T \) in a unit of °C.\(^{20}\)

\[
c_c(T) = 9.99 \times 10^{-7} - 6.94 \times 10^{-5} T^2 + 3.20 \times 10^{-3} T - 2.87 \times 10^{-2}
\]

\[
c_\infty = \text{the vapor concentration in environment temperature } T_c (c_\infty = \rho c (T_c)).
\]

\( r \) and \( R \) are the radial coordinate and contact radius, respectively (see Figure 7a,b). The value of \( r/R \) approaches 1 near the contact line, and \( f(\theta) \) and \( \zeta \) are given by

\[
f(\theta) = (0.27 \theta^2 + 1.30) \left( 0.6381 - 0.2239 \left( \theta - \frac{\pi}{4} \right)^2 \right)
\]

\[
\zeta = \frac{(\pi - 2 \theta)}{2(\pi - \theta)}
\]

In our analysis, temperatures at the contact line are recorded as \( T_{C,L} \) for the left side and \( T_{C,R} \) for the right side. The two temperatures come from the measured temperature profile shown in Figure 5a,c. They are used by eq 9 to determine \( u_r \). Top heating at \( t = 180 \text{ s} \) yields \( u_r = 9.3 \text{ mm/s} \) for the left side and \( u_r = 9.4 \text{ mm/s} \) for the right side with \( T_{C,L} = 30.3 \text{ °C} \), \( T_{C,R} = 30.5 \text{ °C} \), \( \theta = 0.961 (55.1^\circ) \), and \( R = 1.05 \text{ mm} \). The almost equal radial velocities on the two sides explain the uniform coffee-ring profiles along the circumference. For side heating, the left side and right side represent the sunny side and night side, respectively. At \( t = 180 \text{ s} \), the measured parameters \( \theta = 1.095 \text{ °C} \) and \( R = 1.09 \text{ mm} \) with two different temperatures \( T_{C,L} = 32.0 \text{ °C} \) and \( T_{C,R} = 28.9 \text{ °C} \) create \( u_r = 2.0 \text{ mm/s} > u_r = 1.6 \text{ mm/s} \), explaining enhanced nanoparticle deposition on the sunny side than on the night side to display asymmetry characteristic.

The surface temperature gradient in CLR causes Marangoni flow, characterized by Marangoni number \( Ma \)

\[
Ma = \frac{\sigma \chi \Delta T}{\mu \sigma}
\]

where \( \chi \) is the arc length corresponding to \( \Delta T = T_{C,L} - T_{C,R} \) based on measurements (see eq 8 for definition), \( \sigma \) is the surface tension gradient, which is a physical property of water \((\sigma = 1.657 \times 10^{-4} \text{ N/(m K)}) \), \( \mu \) is the dynamic viscosity of water \((\mu = 1.005 \times 10^{-3} \text{ Pa s}) \), and \( \sigma \) is the thermal diffusion coefficient of water \((\sigma = 1.4 \times 10^{-7} \text{ m}^2/\text{s}) \). The estimation reaches \( Ma = 620 \) at \( t = 180 \text{ s} \) for top heating. For side heating, the larger \( \chi \) and \( \Delta T \) creates larger \( Ma = 2303 \) on the sunny side than \( Ma = 611 \) on the night side, \( Ma > 100 \) ensures well-established Marangoni flow in CLR.\(^7\) Marangoni flow helps circulate and confine nanoparticles in CLR. For side heating, the stronger Marangoni flow enhances heat transfer between nanoparticles and base fluid on the sunny side compared with the night side.

Multiscale Energy Analysis in the Droplet. Top heating generates uniform irradiation along the droplet circumferential direction. Here, we explain how the side heating induces higher temperatures on the sunny side. The total heat transfer rate \( Q \) is divided into infrared energy absorption \( Q_{IR} \) and electromagnetic energy absorption \( Q_{EL} \) in a visible band. \( Q_{EL} \) is given by

\[
Q_{EL} = Q - Q_{IR}, \quad Q = \rho_l h_v \frac{dV}{dt}, \quad Q_{IR} = \rho_l h_v \frac{dV}{dt}
\]

\[
c_\text{IR} = \text{the latent heat of vapor evaporations of pure water and nano fluids not absorbed by the nanoparticle.}^2 \text{ The } V^{-c} \text{ curves at different } c_\text{values can be seen in Figure S5 of the Supporting Information.}
\]

At \( t = 180 \text{ s} \), the results are \( Q = 1.37 \times 10^{-2} \text{ W} \), \( Q_{IR} = 1.08 \times 10^{-2} \text{ W} \), and \( Q_{EL} = 2.91 \times 10^{-3} \text{ W} \) for top heating, but \( Q = 9.25 \times 10^{-3} \text{ W} \), \( Q_{IR} = 7.99 \times 10^{-3} \text{ W} \), and \( Q_{EL} = 1.26 \times 10^{-3} \text{ W} \) for side heating. A larger \( Q \) for top heating corresponds to shorter droplet lifetime than side heating (see Figure 3). One notes that infrared energy absorption occurs in a thin liquid film,\(^1^2\) which is marked as region I in Figure 7c,d. Side heating generates infrared energy absorption covering 86.4% of the total energy absorption. Such energy is absorbed in region I of the sunny side only, accounting for one of the reasons to have higher temperatures on the sunny side than on the night side. However, region II in a larger length scale does not receive infrared energy at all (see Figure 7c,d).

Now, \( Q_{EL} \) assigned in CLR and BVR is analyzed. Because nanoparticles are closely packed with each other in CLR, \( Q_{EL, CLR} \) is difficult to be determined. Thus, we turn to calculate \( Q_{EL, BVR} \) using the Lambert–Beer law and Mie scattering theory.\(^{18,30} \) The computation model involves a projected droplet area \( \Omega \) with the microprojected area recorded as \( dS \) (see Figure 7). An original point \( O' \) is defined as the droplet surface corresponding to \( dS \). Optical length \( y \) is the distance from \( O' \) to \( dS \). Nanoparticles receive the radiation flux density \( I_{\omega} \) in the visible region as

\[
I_{\omega} = I_0 (1 - e^{-k_{IR} y})
\]

where \( I_{\omega} \) is the incident radiation flux density at \( \lambda \). For a specific \( d_{ir} \), \( I_0 \) is scaled by the spectrum curve in Figure S1 of the Supporting Information. \( k_{IR} \) is the spectral extinction coefficient\(^1^3\)

\[
k_{IR} = \frac{6 \pi \rho_{IR} d^5_{IR}}{\pi \rho_d d^5}
\]

where \( \rho_p \) and \( d_p \) are the gold nanoparticle density and diameter, respectively. \( \sigma_t \) is the extinction cross section of gold nanoparticles obtained by the Mie scattering theory.\(^1^8 \) \( c_{BVR} \) is the time-dependent nanofluid concentration in BVR. \( k_{fl} \) is time-dependent due to varied \( c_{BVR} \) versus time. The heat generation of electromagnetic energy in BVR is given by

\[
Q_{EL, BVR} = \int_{\lambda=300nm}^{900nm} \int_{\Delta=300nm}^{\infty} I_{\omega}(1 - e^{-k_{IR} y}) dS d\lambda
\]

Equation 17 needs \( c_{BVR} \) to determine \( Q_{EL, BVR} \)

\[
c_{BVR} = \frac{(n - n_{CLR}) h_v V_p}{V - V_{coffee ring}}
\]

where \( n \) is the total number of nanoparticles, determined by \( c_i \) and \( V, V_p \) is the single nanoparticle volume. We note that both the coffee ring and island can accumulate nanoparticles. Here,
the number of particles in CLR \( (n_{CLR}) \) is directly decided by the measured coffee-ring profile (see Figure 6). Equation 18 is correct based on an approximation that nanoparticle deposition occurs at the beginning of light irradiation. This can be explained by Strouhal number \( Sr = u e / R \), which is the ratio of droplet lifetime \( e \) to nanoparticle traveling time from the droplet center to the contact line via radial flow. For an example case with \( e = 600 \) s shown in Figure 3b, \( u e \) equals 0.022 mm/s using eq 9 at the early time of droplet evaporation. Thus, \( Sr = 13.2 \) concludes that nanoparticle deposition occurs in the timescale one magnitude shorter than \( e \).

Calculations are performed at \( t = 180 \) s with \( c_1 = 50 \) ppm, yielding \( Q_{EL, BVR} = 1.9 \times 10^{-6} \) W and \( Q_{EL, CLR} \approx Q_{EL} = 2.91 \times 10^{-3} \) W for top heating and \( Q_{EL, BVR} = 9.15 \times 10^{-7} \) W and \( Q_{EL, CLR} \approx Q_{EL} = 1.26 \times 10^{-3} \) W for side heating. Even though CLR is three magnitudes smaller than the droplet, it dominates all the visible energy absorption. Besides, side heating results in \( Q_{EL} \) being smaller than top heating by 56.7%, indicating the stronger shadow effect for side heating.

A recent study shows that there exists a critical nanoparticle concentration \( c_1 > 10 \) ppm creates extra nanoparticles underneath effective nanoparticle layers. The hidden nanoparticles are not useful for plasmonic heating. Attention is paid to the shadow effect for side heating. Because light irradiates coffee-ring particles along the horizontal plane, effective layers and hidden layers are consecutively arranged along the horizontal direction on the sunny side. Effective layers are exposed to light, but hidden layers are at the back of effective layers. This means that the total visible energy is totally consumed by the sunny side. Night side particles are thoroughly shadowed by sunny side particles (see Figure 7d). Thus, for side heating, the visible energy is almost received in CLR of the sunny side only. In one word, the sunny side not only contributes dominant infrared energy absorption but also dominates the visible energy absorption, yielding higher temperatures than the night side.

In this paper, we use a solar simulator, whose spectrum is almost identical to that of sunlight. The visible energy covers 43% of the light energy over the whole wavelength band range. Our analysis indicates that 31.6 and 51.2% of the incident visible energies are absorbed by droplets for side heating and top heating, respectively. The plasmonic heating can be enhanced by using core–shell nanoparticles, graphene, or nonspherical nanoparticles to raise visible energy absorption. The energy efficiency for droplet evaporation is defined as

\[
\eta = \frac{Q}{Q_{IR,i} + Q_{EL,i}} = \frac{Q}{Q_{IR,i}(1 + \omega)} = \frac{Q}{Q_{IR}(1 + \omega)}
\]

(19)

where \( Q_{IR,i} \) and \( Q_{EL,i} \) are incident light energies in the infrared band and visible band, respectively, and \( \omega = Q_{EL,i}/Q_{IR,i} \approx 0.754 \) for solar energy. Equation 19 shows that all the infrared energy is absorbed \( (Q_{IR,i} = Q_{IR}) \), but only part of visible energy can be received. The efficiency \( \eta \) is 57% for pure water droplet evaporation but increases to 66.0% for side heating and 72.3% for top heating when using nanofluid. The larger efficiency by top heating is because the projected area of light is constant for top heating but decreases for side heating.

**Effect of Various Parameters on Side Heating-Induced Droplet Evaporation.** The above sections focus on the analysis of light heating-induced droplet evaporation with fixed \( V_i = 2.0 \pm 0.1 \) µL and \( q_f = 1400 \) W/m². Effects of \( V_i \) and \( \theta_i \) on droplet evaporation are presented in Figures 8—10, where \( \theta_i \) is the initial contact angle. When a hydrophilic wall is used, droplet dynamics, surface temperatures, and the coffee ring display similar characteristics, as shown in Figures 8—10.
Figure 10. Effect of \( \theta \) on droplet dynamics for side heating with \( q_e = 1400 \text{ W/m}^2 \) and \( c_i = 50 \text{ ppm} \): (a–c) droplet evaporation dynamic on hydrophilic wall and hydrophobic wall; (d) nanoparticle pattern on the hydrophobic wall. Droplet evaporation on hydrophilic wall displays a linear \( V-t \) curve, CCD mode, and nonuniform particle deposition in the coffee ring. Droplet evaporation on the hydrophobic wall displays a nonlinear \( V-t \) curve and a consecutive “stick–slip” feature. Different stages yield different contact lines. Nanoparticles deposit in the last stage as a circular shape.

Regarding the \( q_i \) effect, \( q_i = 0, 800, \) and 1400 W/m² are used (see Figure 9). With the increase in \( q_i \), both the droplet evaporation rate and variation speed of contact angles become larger. Droplet surface temperatures are higher at the same time for larger \( q_i \). During evaporation, part of nanoparticles are deposited on the coffee ring, and the residual particles are left on the island. The increase in \( q_i \) from 800 to 1400 W/m² widens the coffee-ring cross section. This is because nanoparticle deposition on the coffee ring is accelerated caused by enhanced internal flow, causing deposition of fewer nanoparticles on the island.

Three differences exist with \( \theta = 80.2^\circ \) for hydrophilicity and 114.5° for hydrophobicity (see Figure 10). First, the droplet on the hydrophilic wall yields linear evaporation of \( V-t \) but, on the hydrophobic wall, displays nonlinear evaporation characteristic (see Figure 10a). Second, a single constant contact diameter (CCD) is ensured on the hydrophilic wall. However, a droplet lifetime includes multi-CCD stages on the hydrophobic wall. The end of each stage is marked by \( t_1, t_2, t_3, \) and \( t_4 \) (see Figure 10b,c). The transition from a CCD stage to a new CCD stage is paid attention here. During each stage, the residual surface energy of droplet (\( \Delta U_e \)) increases with decreasing \( \theta \). When \( \theta \) decreases to a specific value, \( \Delta U_e \) begins to exceed the pinning energy barrier, leading to a quick receding of the contact line. Thus, a new CCD stage begins at an increased \( \theta \) and a smaller \( D \). The consecutive CCD stages result in the “stick–slip” mode, which is also found for substrate heating. Third, droplet evaporation on the hydrophilic wall produces a coffee ring. Alternatively, several coffee-ring circles having a few nanoparticles are observed on the hydrophobic wall. Each circle corresponds to a CCD stage. Most of nanoparticles are deposited on the center island whose diameter corresponds to the final CCD stage (see Figure 10d).

Wall roughness influences droplet evaporation via wall wettability. For the hydrophilic wall, hydrophilicity is enhanced by raising wall roughness, further ensuring the CCD mode. For the hydrophobic wall, the wall becomes more hydrophobic with larger roughness to yield the “stick–slip” mode. The top light heating-induced droplet evaporation on a superhydrophobic wall with \( \theta = 157^\circ \) is demonstrated in Figure S6 of the Supporting Information. CCA mode is maintained with decreased \( D \). After droplet drying, nanoparticles are observed on the substrate in a local region of 103 \( \mu \text{m} \), noting the initial contact diameter of 650 \( \mu \text{m} \). The CCA mode can be considered as a limit of the “stick–slip” mode when the duration time of each CCD stage approaches zero.

As mentioned in Introduction, different heat sources result in different flow/temperature fields in the droplet. In this paper, the solar simulator contains a mixed wavelength band including visible fraction and infrared fraction. Again, the case with \( q_e = 1400 \text{ W/m}^2, V_i = 2.0 \text{ \mu L}, \) and \( c_i = 50 \text{ ppm} \) is taken as an example for side heating. Even though the visible energy

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only accounts for \(~14\%\) of the total energy absorption, such energy absorption occurs in CLR, whose volume is two to three magnitudes smaller than the whole droplet. The strongly local plasmon heating results in higher temperatures near the contact line than elsewhere. On the contrary, infrared light-induced droplet evaporation causes different temperature distributions. One notes that infrared energy absorption covers the whole droplet surface. The local heating effect near contact line does not exist. Thus, the infrared irradiation results in higher temperature at the apex and lower temperature at the contact line.

Now, the asymmetric light-driven and substrate heating-induced droplet evaporation are discussed. First of all, asymmetric light heating results in two length scales of BVR and CLR, referring to a larger droplet scale and microscale near the contact line, respectively. Radial flow is established in BVR to transport nanoparticles from the droplet center to the contact line. Because radial flow is stronger at the sunny side than the night side, more nanoparticles are deposited at the sunny side. The temperature gradient is weak along the droplet surface corresponding to BVR but becomes significant in CLR. Thus, Marangoni flow is only confined in CLR. Alternatively, for asymmetric substrate heating, the temperature of the free surface decreases monotonically from the hotter side to the colder side. The multiscale feature does not exist. Specifying higher substrate temperature at the left and lower temperature at the right, a clockwise circulating flow is established in the droplet. Such bulk flow transports particles from the hotter side to the colder side to deposit particles in a local region, which is totally inverse to the particles deposition behavior studied in this paper.

**CONCLUSIONS**

Solar-driven droplet evaporation is investigated. By correcting the IR energy emission from the substrate near the contact line, we achieve precise measurement of droplet surface temperatures. It is shown that solar-driven droplet evaporation does display the nonmonotonic temperature variation, guiding us to decouple the droplet into a contact line region (CLR) and a bulk volume region (BVR). The steep temperature gradient in CLR and flat distribution in BVR are recognized as the boundary layer effect. Radial flow in BVR transports nanoparticles from the droplet body to the contact line, while Marangoni flow is only confined in CLR. The coffee-ring nanoparticle deposition dominates the visible energy absorption, explaining higher temperatures in CLR than elsewhere.

Top heating demonstrates symmetric evaporation characteristic. For side heating, the sunny side holds stronger radial flow to deposit more nanoparticles on the substrate than the night side. Furthermore, the light shadow effect results in weak heat generation of nanoparticles on the night side. Thus, the sunny side has higher contact line temperatures than the night side, indicating asymmetric behavior. For the example case of \(V_1 = 2.0 \mu \text{L}, q_e = 1400 \text{ W/m}^2\), and \(c_i = 50 \text{ ppm}\), the energy absorption efficiency of light is 66.0% for side heating and 72.3% for top heating. The projected area of light is constant for top heating but decreases for side heating, resulting in the efficiency difference between the two irradiation directions.

On the hydrophilic wall, evaporation rates and variation speeds of contact angles may be altered when initial droplet volumes and incident irradiation fluxes are changed, but the conclusions are the same. The evaporation mode of constant contact diameter (CCD) on the hydrophilic wall is switched to the “stick–slip” mode on the hydrophobic wall due to insufficient pinning energy needed to overcome the residual surface energy of liquid during evaporation.

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.langmuir.9b03122.

Calculation method of \(I_i/I_0\) versus \(I_0\); the major parameters, instruments, and uncertainties; the spectrum of xenon lamp compared with the sun spectrum; characterization of nanoparticles and nanofluid; the transmittance spectrum of liquid with an optical length of 200 \(\mu\text{m}\) in the wavelength range of 3–5 \(\mu\text{m}\); curve fitting equation for droplet morphology; \(V−t\) of droplets for top heating and side heating; and top light irradiation induced droplet evaporation on superhydrophobic wall

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**Notes**

The authors declare no competing financial interest.

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