Steady State Vapor Bubble in Pool Boiling

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Boiling, a dynamic and multiscale process, has been studied for over five decades; however, a comprehensive understanding of the process is still lacking. The bubble ebullition cycle, which occurs over millisecond time-span, makes it extremely challenging to study nearsurface interfacial characteristics of a single bubble. Here, we create a steady-state vapor bubble that can remain stable for hours in a pool of sub-cooled water using a femtosecond laser source. The stability of the bubble allows us to measure the contact-angle and perform *in-situ* imaging of the contact-line region and the microlayer, on hydrophilic and hydrophobic surfaces and in both degassed and regular (with dissolved air) water. The early growth stage of vapor bubble in degassed water shows a completely wetted bubble base with the microlayer, and the bubble does not depart from the surface due to reduced liquid pressure in the microlayer. Using experimental data and numerical simulations, we obtain permissible range of maximum heat transfer coefficient possible in nucleate boiling and the width of the evaporating layer in the contact-line region. This technique of creating and measuring fundamental characteristics of a stable vapor bubble will facilitate rational design of nanostructures for boiling enhancement and advance thermal management in electronics.

Boiling is one of the most efficient heat transfer mechanisms that allows a large amount of heat to be transferred over small surface areas due to the associated phase-change processes. It has been widely used in industry¹, from cooling small electronics to large power plants. However, it is a complex phenomenon that involves multiple length and time scales that are present at the base of a bubble in the contact line region^{2,3}. The three-phase contact line region (Fig. 1a), where the liquid-vapor interface meets the solid surface, can be divided into three sub-regions of varying thicknesses: non-evaporating film region (of nanometer-scale thickness), evaporating film region (of micrometer-scale thickness), and bulk meniscus region (of micrometer- to millimeter-scale thickness)^{4,5}, with these regions constituting the microlayer (Fig. 1b). Contact line models^{6,7}, together with transient conduction⁸ and microlayer evaporation^{9,10}, have been widely accepted as the basic heat-transfer mechanisms in boiling. The dynamics of contact line region and the microlayer dictate bubble growth and departure, and are of significant importance in understanding the fundamental behavior of the boiling phenomenon¹¹. Visualization of the boiling process and the contact line region has recently been pursued¹²⁻¹⁷ and had tremendous impact in providing a realistic depiction of the boiling process; however, the unsteady nature and a short time-span of the bubble ebullition cycle has made *in-situ* imaging of a single bubble very challenging.

The contact line region is incorporated into predictive boiling models through contact angle values¹⁸⁻²⁰. The intricacies involving the shape of an interface and the behavior of the contact line are implicitly accounted for in the contact angle, thus making the bubble contact angle parameter of significant importance in boiling models. Methods that have been applied to determine the contact angle include the captive-bubble technique (involves an air-bubble)²¹⁻²³, the flotometric technique (involves solid particles interacting with a bubble)²⁴, and high-speed

photography (involves visual approximation of the contact line region and angle)²⁵. In addition, equilibrium or advanced (receding) contact angles of a liquid droplet on the surface at room temperature are often used for boiling models^{19,26}, although it is difficult to relate the boiling process to the droplet wetting characteristics due to the highly transient conditions associated with liquid-vapor phase change²⁷⁻²⁹. Thus, the contact angle of a vapor bubble in pool boiling has yet to be measured in the early growth stages due to the dynamic nature of the bubble ebullition cycle; making it all the more necessary to image the contact line region to advance the understanding of boiling process and enhance boiling heat transfer efficiency.

In this work we create a steady-state vapor bubble, in a pool of sub-cooled water, by heating using a femtosecond laser source. The bubble remains stationary for hours, allowing *in*situ imaging of the microlayer and the contact line region, and measurements of the contact angle. A Ti:Sapphire ultrafast laser (pulse length ≈ 120 fs, repetition rate = 80 MHz, center wavelength $\lambda_0 = 800$ nm) in conjunction with a second-harmonic generation (SHG) unit was used to generate high-power laser pulses at a free-space wavelength of $\lambda_{SHG} = 400$ nm. The laser pulses were passed through a $5 \times$ or $50 \times$ objective lens and focused on an absorbing 40 nm thick Au film that is sandwiched between a silica glass substrate (bottom-surface) and sputter deposited 400 nm thick layer of SiO_2 (top-surface). The focused laser beam creates a highly localized heating area corresponding to the beam-diameter $\approx 170 \ \mu\text{m}$. To achieve boiling, a pool of water was created inside a 6 cm long and 1.4 cm inner diameter glass tube bonded to the SiO₂ top-surface of the substrate. Although laser-initiated bubbles have been used in literature $^{30-32}$, fundamental characteristics of formation of such bubbles or their *in-situ* imaging to understand the boiling process have not been explored. Experiments were performed in both regular deionized (DI) water with dissolved air and degassed DI water. The latter was prepared by

boiling regular DI water for one-hour, and filling it inside the glass-tube using a 220 nm filter syringe to remove any particulates. Based on the one-dimensional (1D) diffusion equation, it would take > 24 hours for the air to diffuse to the bottom of the 6 cm long tube whereas each experimental measurements lasted for < 2 hours. Convection currents due to bubble formation can increase diffusion of gases in water; however, experimental observations show that water remained degassed near the surface as the vapor bubble condensed when the laser was turned off (video V1 in the supplementary material). Using regular DI water, we tested hydrophilic SiO₂ surfaces (where experiments were performed immediately after plasma cleaning) and normal SiO₂ surfaces. Similarly, using degassed DI water, we tested both the normal SiO₂ surface and a hydrophobic tridecafluoro-1,1,2,2-tetrahydrooctyl-trichlorosilane (FOTS) surface. In Table I, we list the drop contact angle on these surfaces measured using a goniometer (drop images in supplementary information). After a bubble is formed on the surface through heating with a laser pulse, we increase the average power of the laser by 20 mW for every subsequent reading of the bubble base, bubble diameter and contact angle. The measurements were stopped before the average laser power could reach the damage threshold of the SiO₂ surface (corresponding to an average power ≈ 240 mW). The uncertainty in all the contact angle measurements are one standard deviation for repeated experimental measurements (five in total). Please refer to the supplementary information for details on the experimental setup, sample fabrication and preparation.

Figures 2a, 2b and 2c show the bottom view, obtained using an inverted optical microscope, of a single bubble formed on hydrophilic SiO₂, FOTS and normal SiO₂ surfaces, respectively, with increasing laser heating power. The vapor bubble instantly achieves steady-state in degassed DI water as the heat transfer from the surface leads to continuous evaporation

of water in the microlayer which is balanced by the continuous condensation of vapor at the liquid-vapor interface away from the surface due to the sub-cooled pool of water (temperature of water was ≈ 75 °C lower than the saturation temperature ≈ 100 °C). The bubble contact angle θ for the hydrophilic SiO₂ surface was obtained by measuring the bubble base diameter, d_{bb} , and the height of bubble middle plane, z_1 , (Fig. 2-d1) and determined by the equation: $\theta =$ $\arctan(d_{bb}/2z_1)$; whereas for the hydrophobic surface (FOTS), θ was obtained by the firstorder derivative of the parabolic curve of the interface (Fig. 2-d2) and given by $\theta = \pi + \pi$ $\arctan(-4z_2/d_{bb})$ where the height of the bubble is z_2 . In these equations, the bubble base diameter, d_{bb} , was obtained directly from the calibrated optical images acquired using a CCD camera; while the z positions of the bubble middle plane (for hydrophilic and normal SiO_2) or the bubble height (for the FOTS surface) were obtained by translating the focal plane of the objective to the appropriate z height and reading the z-offset from the controller of the motorized translation stage. Figs. 2e and 2f show the variation in the bubble base/bubble diameter and contact angle, respectively, with increasing laser power for the various cases studied. The bubble sizes were consistently smaller in degassed DI (D) water when compared to regular DI (R) water due to the contribution of dissolved air in the bubble growth phenomenon in regular water. This effect was further confirmed by turning the laser off; the bubble in degassed DI water disappeared in < 20 seconds (due to condensation of vapor) while the bubble in regular DI water decreased in diameter slightly but stayed on the surface for days (videos V1, V2 and V3 in the supplementary material). Our results are also consistent with a recent study³³ where air nanobubbles were found to be stable for days due to the slow-rate of dissolution of air into an already saturated surrounding liquid.

For the normal SiO₂ surface with degassed water, the bubble contact angle decreased with increasing laser power (from 73.6° ± 3.9° at 120 mW to 45.3° ± 5.2° at 200 mW). In all the other cases, the bubble contact angle was found to be independent of the laser power studied. Average bubble contact angle in regular water was determined to be $31.9^{\circ} \pm 0.5^{\circ}$ on normal SiO₂ surface, which is similar to the drop contact angle (Table I), and in good agreement²⁶ with the drop receding contact angle after boiling experiments ($32.3^{\circ} \pm 0.4^{\circ}$). The average contact angle was $29.3^{\circ} \pm 0.4^{\circ}$ on the hydrophilic SiO₂ surface which is slightly smaller than the bubble contact angle on normal SiO₂ surface. Contact angle of the bubble on FOTS surface was $96.8^{\circ} \pm$ 0.2° which is also similar to the measured drop contact angle. These variations in contact angle, especially between degassed and regular water on the same surface, depend on the dynamics of the microlayer and contact line region as studied and explained below.

A stable bubble enables *in-situ* imaging of the contact line region present at the base of the bubble. Using femtosecond laser illumination through a 50× microscope objective, bubbles were formed on a normal SiO₂ (Fig. 3a) surface with regular water, a normal SiO₂ surface with degassed water (Fig. 3b), and a FOTS surface with degassed water (Fig. 3c). The contact line region is imaged (Fig. 3) using the inverted optical microscope under illumination from both a white halogen lamp source (Figs. 3-a1, b1, c1) and a 632 nm HeNe laser (Figs. 3-a2, b2, c2). The microlayer and the contact line region were identified based on the juxtaposition of these two sets of images. Under coherent HeNe laser illumination, two sets of fringes were observed in the images that are a result of thin-film interference associated with interaction between regions of different refractive index. The first set of fringes, F-1 (dark thick partial rings), have fringe-gaps decreasing in the outward radial direction and are associated with interference resulting from the top curved interface of the bubble (and not due to the contact line region). The second set of

fringes, F-2, are relatively closely packed and the fringe-gap for these set of fringes increase in the outward radial direction. These fringes are a result of thin-film interference of incident light with the partially reflected light within the thin liquid microlayer present at the base of the bubble,¹⁴ with the increase in fringe-gap attributed to the increase in radius-of-curvature of the microlayer in the outward radial direction. The second set of fringes is clearly evident in degassed DI water (Figs. 3-b2, c2) showing the presence of a liquid microlayer over the entire bubble base. However, in regular DI water (Fig. 3-a2), the fringes are absent from the center of the bubble base and are only present in the equivalent bright regions of Fig. 3-a1. This observation implies the presence of a dry-spot region at the center of the bubble base and the formation of the three-phase contact line region (liquid-vapor-solid) interfacing with the SiO₂ surface, with a significantly reduced microlayer. The microlayer shapes obtained for a bubble on normal SiO₂ in regular water and degassed water, and on FOTS surface in degassed water is plotted in Figs. 2-a3, b3 and c3 respectively. As the interference of the monochromatic light source generates dark and bright fringes corresponding to constructive and destructive interference respectively, these fringes are separated by an optical path difference equal to effective half wavelength, $n\lambda_0/2$, where n is the refractive of the medium, λ_0 is the free-space wavelength of light. The position of these fringes is used to construct the shape of the microlayer, where the difference in local thickness at the adjacent bright/dark fringe location t_{m+1} and t_m is given by $t_{m+1} - t_m = \lambda_0/2n \cos(\theta)$ for the light refracted at angle θ into the microlayer.

The fringes observed in the contact line region are also used to explain the experimentally measured contact angle values. In the regular DI water on normal SiO_2 surface, the larger size of the bubble (due to contribution of dissolved air) at low laser power creates a dry spot at the center causing the creation of a three-phase contact line; and hence, the bubble contact

angle is similar to the drop contact angle (where a similar three-phase contact line is present). However, with degassed DI water on normal SiO₂ surface, the microlayer covers the entire bubble base preventing the formation of the three-phase contact line, and the contact angle is governed by the microlayer curvature relative to the bubble curvature. Hence, the contact angle decreases with increasing laser power as the radius of curvature of the microlayer increases significantly faster compared to the radius of curvature of the bubble. Similarly in FOTS, the larger radius of curvature microlayer along with the parabolic bubble shape results in large contact angle values. The parabolic shape of bubble is attributed to the larger bubble base diameter as the reduced wettability of the hydrophobic surface requires a larger microlayer to remove the same amount of heat from the surface. However, for degassed DI water on both normal SiO₂ and FOTS surfaces, it is expected that after a critical bubble size is reached – the microlayer would reduce in thickness, form the three-phase contact line and the bubble contact angle would converge to that of the drop contact angle.

Further, bubbles with the microlayer wetting the entire bubble base will not depart the surface as the capillary and disjoining suction force (due to reduced liquid pressure in microlayer) is estimated to be larger than the buoyancy force and capillary force at the top of the bubble (please refer to supplementary information for detailed analysis).

Maximum heat flux occurs in the thin evaporating region¹⁰ and is of critical importance in bubble growth dynamics; however, knowledge of heat transfer coefficient and corresponding width of this region is currently lacking in literature. We use experimental data from *in-situ* imaging of the contact line region together with finite-element-method based numerical simulations to characterize the evaporating region in the microlayer. We first focus on the bubble formation in regular DI water on normal SiO₂ surface to obtain experimental data. Interestingly,

it was found that the bubble grew gradually at constant laser power. The source of bubble growth results from the air dissolved in the water, which is released into the bubble during the vaporization of water from the evaporating region of the microlayer. The bubble grew steadily (Fig. 4a) at a volumetric rate of $(5.60 \pm 0.06) \times 10^{-3}$ mm³/min (please refer to supplementary information for bubble volume calculations) and the contact line region at the bubble base grew radially outward at a speed of (1.9 ± 0.1) µm/min during the initial 40 min, but stopped after it reached a diameter of $\approx 270 \,\mu m$ (Fig. 4b); this limiting diameter approximately corresponds to the measured laser beam diameter ($\approx 170 \ \mu m$) with additional radial heat conduction in the Au layer. The uncertainty in the measurements of the bubble growth rate and bubble base are standard deviation of the fit parameter. Contact angle of the bubble (Fig. 4c) decreased with time as the bubble base remained nearly constant while the bubble diameter grew uninhibited. Similar to Fig. 3-a1, the central dry spot diameter was identified from *in-situ* imaging of the contact line region. Based on these experimental data, the heat transfer rate q in the evaporating region can be obtained from the air-water solubility mass balance calculation: $q = \dot{m}_{water} \Delta H =$ $(\dot{m}_{air}/S_a)\Delta H$, where \dot{m}_{water} is the vaporization rate of water in evaporating region, \dot{m}_{air} is the mass flow rate of air into the vapor bubble from evaporating region, S_a is the solubility of air in

water, and ΔH is the latent heat of vaporization.

The heat transfer rate in the evaporating region q is also dependent on the overall heat transfer coefficient h and the area of the evaporating region through (please refer to supplementary information): $q = h(\pi D_{bb}w)\Delta T$ where D_{bb} is the central dry spot diameter, w is the width of evaporating region, and ΔT is the temperature difference between the surface and the bulk fluid. Unknown parameters h and w characterize the evaporating region (Fig. 4d), and we performed finite-element-method based simulations to determine the range of h and w for

which the simulated release rate of air from the evaporating region agreed with that obtained through measured bubble geometry in the experiments (Fig. 4a). An axi-symmetric domain was considered that included the glass substrate, 40 nm Au layer and 400 nm SiO₂ layer (Fig. 4d). A parametric study was performed where h and w were varied from 5000 $Wm^{-2}K^{-1}$ to 200,000 Wm^{-1} 2 K⁻¹, and from 0.5 µm to 19.5 µm, respectively for a total of 3500 simulation cases. Figure 3e shows the range of h and w for which simulation results were in good agreement with experiments within a standard uncertainty of 4.5 %. The temperature profile of the surface is plotted for this range (Fig. 4f). Interestingly, the surface temperature at $r \approx 135$ um was ≈ 39 °C. which is the critical temperature when Marangoni flow inhibits fluid flow towards the contact line³⁴, thus equilibrating the incoming mass flow to the evaporation rate and causing the contact line to become stable at bubble base diameter of $\approx 270 \ \mu m$. The temperature at the center of the bubble is calculated to be ≈ 82 °C, which is also in good agreement with experiments³⁵, where it has been shown that the formation of an air bubble in pool boiling in sub-cooled water at room temperature occurs at ≈ 84 °C. The thermal boundary layer thickness prior to bubble nucleation is simulated to be $\approx 200 \,\mu\text{m}$, and around the steady bubble is estimated to be $\approx 280 \,\mu\text{m}$ for h = 120kW/m²K and $w = 10 \mu m$ (please refer to supplementary information).

In summary, a steady-state vapor bubble is created in a pool of sub-cooled water by femtosecond laser heating which allows for *in-situ* imaging of the microlayer and the contact line regions. The bubble can remain stable for hours as the evaporation of water at the surface is balanced by condensation of vapor at the liquid-vapor interface inside the bubble. Experiments are conducted on hydrophilic (SiO₂) and hydrophobic (FOTS) surfaces in regular (with dissolved air) and degassed DI water. The contact angle of the vapor bubble is measured for various cases, and the microlayer and contact line region are imaged with white light and a coherent laser

source. For the laser powers studied, it was found that the three-phase contact line readily forms in regular DI water, while the microlayer covers the entire bubble base in degassed DI water. The contact angle for the bubble is found to resemble the drop contact angle on the same surface if the three-phase contact line forms, otherwise the contact angle is dependent on the curvature of the microlayer and the bubble, and decreases with increasing laser power. The evaporating region in the contact line region is characterized by numerical simulations and experimental results, and permissible values of heat transfer coefficient and corresponding width are calculated, thus providing an estimate to the upper limit of the heat transfer coefficient attainable in nucleate boiling as well as thin-film evaporation. The work presented here will advance the design of nanostructures to enhance heat transfer by optimizing the width of microlayers, and improve our understanding of boiling phenomenon in outer-space where lack of gravity causes the bubbles to stay stationary on a heated surface. *In-situ* imaging of the microlayer and contact line region in a steady state bubble is a powerful technique for understanding the physical dynamics of the bubble growth process.

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Author Contributions

A.A. and S.C.M. conceived the ideas and directed the work. S.C.M., A.Z., A.C. and A.A. designed the study. A.Z., A.C. and S.C.M. carried out the experiments and analyzed the data with assistance from A.A and P.C.W. A.Z. performed the numerical simulations with inputs from S.C.M., A.C., A.A and P.C.W. S.C.M., A.A., A.Z., and A.C. wrote the manuscript.

Competing Financial Interests

The authors declare no competing financial interests.

Figure Legends

Figure 1: Microlayer and three-phase contact line evolution during a vapor bubble growth process. (a) Three-dimensional (3D) schematic of a vapor bubble on a heated surface in a pool of liquid depicting the presence of the three-phase contact line. Cross-sectional two-dimensional (2D) view of the three-phase contact line showing the non-evaporating region (NER), evaporating film region (EFR) and bulk meniscus region (BMR); microlayer includes EFR and part of the BMR. (b) 2D schematic of vapor bubble on a heated surface with a microlayer over the whole bubble base (three-phase contact line is not present). Cross-section of the base of the bubble depicting the microlayer. Increase in heating temperature and bubble size forms a three-phase contact line with reduced microlayer, *i.e.*, from 1b to 1a.

Figure 2: Laser heated steady-state bubble under 5× magnification captured using an inverted optical microscope on: (a) normal SiO₂ surface in degassed water, (b) FOTS surface in degassed water, and (c) normal SiO₂ surface in regular water, as a function of increasing laser power: 120 mW (a1 and b1), 140 mW (c1), 160 mW (a2 and b2) and 200 mW (a3, b3 and c2). The bubble is twice as large for regular water when compared to the degassed water due to contribution of dissolved air; scale bars in a, b and c are 50 µm. (d) Contact angle measurement depiction on (d1) SiO₂ surface ($\theta = \arctan(d_{bb}/2z)$) and (d2) hydrophobic FOTS surface ($\theta = \pi + \arctan(-4z/d_{bb})$). (e) Bubble base (BB) and bubble diameter (BD) measurements on normal SiO₂ (Norm. SiO₂) surface in degassed (D) and regular (R) water, hydrophilic SiO₂ (Hydro. SiO₂) in regular water, and hydrophobic FOTS in degassed water. (f) Contact angles for bubbles in (f1) regular water and (f2) degassed water with increasing laser power. Bubble contact angles

in regular water were found to independent of laser power and similar to drop contact angles (shown in inset in (f1)), but noticeably larger in degassed water on normal SiO₂ while dropping significantly with increase in laser power. Uncertainties in (e) are based on standard deviation of five measurements for every experimental data point. Uncertainties in (f1) and (f2) are one standard deviation based on propagation of uncertainty from five individual measurements of bubble base and z_1 or z_2 position for each data point. Lines connecting the data points in (e), (f1) and (f2) are guides to the eye.

Figure 3: Optical images of the bubble with $50 \times$ magnification under white-light and HeNe laser illumination. (a) Bubble on normal SiO₂ surface in regular water shows fringes (F-2) only in the corresponding white/bright region but absent at the center implying a central dry-spot and the narrow microlayer forming a three-phase contact line. (b) Bubble on normal SiO₂ surface and (c) FOTS surface in degassed water, show the bubble base is completely covered with the microlayer in bubble base region and no three-phase interfacial line is formed yet. The formation of the three-phase line is expected to change the bubble contact angle to be similar to that of a drop. Secondary fringes F-1 (darker and thicker partial rings) are caused due to the interference of light with bubble curvature as light is incident from the top of the sample. The scale bar in all images is 50 µm. (a3), (b3) and (c3) show the microlayer curvature (shape) obtained from the respective fringes using the thin-film interference equation. (b3) and (c3) do not have a dry spot and the microlayer thickness at r = 0 has been assumed to emphasize this fact. Uncertainties in (a3), (b3) and (c3) for radial distance r are determined from the smallest fringe width that can be measured based on the resolution of the camera.

Figure 4: Bubble growth on normal SiO₂ surface in regular water at constant laser power and corresponding simulation results. (a) Bubble volume growth rate. (b) Bubble base (black) and bubble diameter (red) change with time during growth; for the bubble diameter, the upper and lower limits of the error bar are too close and they merge together. (c) Bubble contact angle change with time during growth. (d) Finite-element-method based simulation domain and boundary conditions. The heat conduction inside the sample was simulated with the 2D axial symmetry condition to estimate the bubble growth rate. A large enough domain (1 mm x 1 mm) was simulated so that the right side boundary condition could be set as thermally insulated. The heat source is the gold layer which absorbs the laser (beam radius $\approx 85 \ \mu m$). The heat transfer coefficient h and width w of the evaporating layer were varied in the simulations, and results for the bubble growth were compared to experimental results, within an error of 4.5 %, to estimate the range of h and w, as depicted in (e). (f) Temperature profile on solid surface from simulations depicting the temperature to be ≈ 39 °C at bubble base radius of $\approx 135 \,\mu\text{m}$. Uncertainties in (a) and (c) are based on propagation of uncertainty from five individual measurements or readings of bubble diameter and contact angle, and bubble base and z_1/z_2 position, respectively, for every experimental data point. Uncertainties in (b) are based on one standard deviation of five measurements for each data point. Rectangular red colored bars in (f) depict the spread in surface temperature for a subset of simulations where bubble growth rate was calculated to be within an error of 0.2 % from experimental results; average temperature values are also shown and connected as a guide to the eye.

Tables

Table I: Sample surfaces and liquids used for the experiments along with the static droplet contact angle on these surfaces. Hydrophilic SiO₂ surface was created by oxygen plasma cleaning of normal SiO₂ surface, and used in experiments immediately afterwards. The normal SiO₂ surface was used approx. 3 to 4 days after oxygen plasma cleaning. Uncertainties in drop contact angle are one standard deviation based on propagation of uncertainty from five individual measurements.

Sample Surface	Drop Contact	Liquid Tested
	Angle	
Hydrophilic SiO ₂	0°	regular DI water
Normal SiO ₂	$33.4^{\circ} \pm 2.7^{\circ}$	regular & degassed DI water
Trichlorosilane (FOTS)	$109.8^{\circ} \pm 2.9^{\circ}$	degassed DI water









Supplementary Information: Steady State Vapor Bubble in Pool Boiling

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S1. Sample Fabrication

Figure S1 shows the cross-sectional schematic of the fabricated samples. A 10 nm thick layer of Cr, followed by a 40 nm thick Au layer and another 10 nm thick Cr layer were thermally deposited onto a silica substrate. The thin metallic layers were used to locally absorb the focused laser beam to create bubbles. A final sputter deposition of 400 nm thick layer of SiO₂ served as the hydrophilic (drop contact angle of 0° immediately after oxygen plasma cleaning) or the normal (drop contact angle recovered to $33.4^\circ \pm 2.7^\circ$ few days after plasma cleaning) SiO₂ surface (Fig. S1a). The FOTS samples were fabricated by molecular vapor deposition of a single monolayer of FOTS on the SiO₂ surface (Fig. S1b).

		FOTS	≈1.3 nm
SiO ₂	≈ 400 nm	SiO ₂	≈ 400 nm
Cr	≈10 nm	Cr	≈10 nm
Au	≈ 40 nm	Au	$\approx 40 \text{ nm}$
Cr	≈10 nm	Cr	≈10 nm
Silica	≈1 mm	Silica	≈1 mm
	(a)		(b)

Figure S1: Cross-sectional schematic of the fabricated samples: (a) top SiO₂ surface (b) top FOTS surface (layer thicknesses not to scale).

S2. Sample Preparation

The final fabricated samples were cleaned with acetone, ethanol and IPA, and rinsed with DI water. After solvent cleaning, the samples were placed in oxygen plasma for 5 minutes. Before every set of experiments, the samples were rinsed with DI water again. The drop contact angle on the fabricated surfaces is measured using a goniometer and images shown in Table S1. The water to be used in the experiments was degassed by boiling it for one hour using hot-plate, and subsequently cooled down to room temperature in a sealed bottle. The liquids (regular or degassed DI water) were passed through a filter with pore size of 220 nm to eliminate any particles or contaminants suspended in the liquid. A pool of water was formed in a 6 cm long and 1.4 cm inner diameter glass tube bonded on the SiO₂ or FOTS surface to achieve boiling. The laser beam illuminated the sample normally through an inverted optical microscope, and the laser power was increased incrementally until the bubble formed. After a stable bubble was formed, the first reading of the stage z position was taken; bubble diameter and bubble base readings were obtained from the calibrated optical images that are simultaneously recorded. The laser power was increased by 20 mW per reading and measurements were stopped immediately before the damage threshold for the sample was reached.

Sample Surface	Drop Contact Angle	Experimental Image of Drop
Hydrophilic SiO ₂	0°	
Normal SiO ₂	$33.4^{\circ} \pm 2.7^{\circ}$	
Trichlorosilane	$109.8^{\circ} \pm 2.9^{\circ}$	
(FOTS)		

Table S1: Drop contact angle on surfaces used in the boiling experiments

S-2

S2.1 Diffusion of Air in Degassed Water

In order to determine the duration during which the degassed water could remain degassed in the vicinity of the SiO₂ surface, the diffusion of air was simplified to a 1-D problem:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2}$$
 Eq. (1)

Eq. (2)

where C(x,t) is concentration of air in water, *D* is diffusivity of air in water, and *t* is time. The following initial and boundary conditions were imposed: (x, 0) = 0 and $C(0, t) = C_0$, where C_0 is the saturation concentration of air in the water at 101.3 kPa and 25 °C.

Solving Eq. (1), the concentration profile as a function of time is shown in Fig. S2 (Eq. 2). Thus, the length of the glass tube was chosen to be 6 cm to keep the water degassed for time scales much longer than the each experiment (approx. t = 1 h).



Figure S2: Diffusion profile of air in degassed water as a function of time to determine the height of glass tube.

S3. Experimental Setup

A Ti:Sapphire ultrafast laser was used to generate femtosecond laser pulses with an average power of 2.7 W at 800 nm (pulse length \approx 120 fs, repetition rate = 80 MHz, center wavelength λ_0 = 800 nm). The laser pulses were then passed through a Second Harmonic Generation unit to generate 400 nm pulses with an average power of 450 mW. The average power of the laser illuminating the sample was controlled by using a continuously variable neutral density (ND) filter. The laser beam was then directed to one of the input ports of an inverted optical microscope. The laser beam was focused onto the sample using a $5 \times$ or $50 \times$ microscope objective. The laser pulses were partially absorbed by the thin metallic layers on the sample and created a stable and highly localized heating area corresponding to the beam size. A 6 cm long glass tube was mounted on the sample to hold the DI water. As the laser power was increased, a nucleation site on the sample surface was formed creating a stationary bubble inside the glass tube. The length of the tube was chosen to prevent diffusion of gasses back into the degassed water to the sample surface where the bubbles are generated. The bubbles were illuminated with a white light halogen lamp and imaged onto a CCD camera through the same microscope objective. To block the residual 400 nm laser beam reflected by the sample, a 425 nm long-pass filter was placed after the beam splitter. Various z-planes of the sample were focused using the motorized control for z-position of the objective (with a z-resolution of approx. 10 nm). The difference between two z-plane readings along with the bubble diameter measurements were used to calculate the bubble contact angle.



Figure S3: Schematic of the experiment setup for bubble contact angle measurement. SHG: Second Harmonic Generator, ND Filter: Continuously variable neutral density filter, HAL: white light halogen lamp for illumination, OBJ: 5× or 50× microscope objective, BS: beam splitter, LPF: 425 nm Long Pass Filter, M: mirror, CAM: 5 megapixel CCD camera.

In order to calculate the power absorbed (P_{ab}) by the sample, a power meter was placed in the path of incident light to measure the input power (P_{in}) . Similarly, the reflected (P_{re}) and the transmitted powers (P_{tr}) were measured. The absorbed power was obtained using Eq. (3) from energy conservation to be $P_{ab} = 0.52P_{in}$.

$$P_{ab} = P_{in} - P_{re} - P_{tr}$$
 Eq. (3)



Figure S4: Schematic representing the incident laser on fabricated sample. The input laser power P_{in} is partially reflected P_{re} and transmitted P_{tr} ; these three variables are experimentally measured to determine the absorbed laser power P_{ab} by the Au layer in the sample.

S4. Bubble Volume

In order to determine the volume of the bubble on the SiO₂ surface, it is divided into two parts (Fig. S5): top part (I) is a hemisphere while the lower part (II) is a partial parabola. Equations for the two sections can be determined by bubble base diameter D_{bb} , bubble diameter D_{bd} and corresponding height *z* of bubble middle plane:

$$y = \frac{1}{4z}x^2 - \frac{D_{bb}^2}{16z}, \qquad x \in \left(-\frac{D_{bd}}{2}, -\frac{D_{bb}}{2}\right) \cup \left(\frac{D_{bb}}{2}, \frac{D_{bd}}{2}\right)$$
 Eq. (4)



Figure S5: Schematic showing a vapor bubble formed on the laser-heated sample in a pool of water. The bubble is approximated as a combination of hemisphere top part (denoted as I) and parabolic lower part (denoted as II). Experimentally measured bubble mid-plane height (z), mid-plane diameter (D_{bd}) and bubble base diameter (D_{bb}) are used to estimate the bubble volume.

The volume of top part I is:

$$V_I = \frac{1}{12} \pi D_{bd}^{3}$$
 Eq. (5)

For an infinite element in lower part II, the volume is $\pi x^2 \cdot dy$. Thus, the volume of part II is:

$$V_{II} = \int_0^z \pi (4zy + \frac{D_{bb}^2}{4}) \, dy = \pi z (2z^2 + \frac{D_{bb}^2}{4})$$
 Eq. (6)

Finally, the bubble volume is the sum of part I and part II:

$$V = \frac{1}{12}\pi D_{bd}^{3} + \pi z (2z^{2} + \frac{D_{bb}^{2}}{4})$$
 Eq. (7)

S5. Finite-Element Numerical Simulations: *h* vs. *w* plot:

In the numerical simulations, the heat transferred through the evaporating region q was expressed as:

$$q = h \cdot \pi \left[\left(\frac{D_{bb}}{2} + w \right)^2 - \left(\frac{D_{bb}}{2} \right)^2 \right] \cdot \Delta T$$
 Eq. (8)

where *h* is the heat transfer coefficient in the evaporating region, D_{bb} is the bubble base diameter, *w* is the width of the evaporating region, and ΔT is the temperature difference between the surface temperature in evaporating region to that of bulk liquid. Eq. (8) can be further simplified to:

$$q = h\pi (D_{bb}w + w^2)\Delta T \qquad \qquad \text{Eq. (9)}$$

 D_{bb} is in the order of 100 µm and w in the simulation is varied from 0.5 µm to 19.5 µm. Thus, the term including w^2 can be ignored making q highly dependent on the product of h and w. As the heat transfer q determines the evaporation rate and bubble volume growth rate, there exists an upper and a lower limit of the product of h and w, the region within which corresponds to a specified error between numerical simulations and experiments.

S6. Thermal Boundary Layer Thickness Estimation

COMSOL simulations are performed to determine the boundary layer thickness δ prior to bubble nucleation (similar to quenching heat flux) and the thickness varies between ~180-270 µm in the region above the heat source. The thermal boundary layer thickness is also estimated around the steady-state vapor bubble to be ~280 µm. Conduction heat transfer in water is assumed as the heat transfer mechanism in the thermal boundary layer.

Quenching heat flux: The domain for COMSOL simulations is described in Fig. S6-a, where the conditions prior to bubble nucleation are simulated. From Fig. 2e of manuscript, it is evident that a bubble does not form until the laser power exceeds \sim 100 mW. Thus, as the laser is turned on gradually from 0 mW, the laser power first heats up the water next to the surface (without bubble nucleation) forming a thermal boundary layer. This process is similar to that seen in traditional boiling methods when the bubble has departed from the surface and the heat flux at the surface is defined as the quenching heat flux which forms the thermal boundary layer. Simulations were performed for two laser powers: 50 mW and 100 mW, for the domain similar to experiments and that adapted in Fig. 4 of the manuscript. The thermal boundary layer thickness variation for these cases is shown in Fig. S6-b, as the temperature varies along the surface. Above the laser beam (heat source) area, the average thermal boundary layer thickness for 50 mW and 100 mW laser powers are found to be ~248 µm and ~195 µm, respectively.



Figure S6: (a) Schematic of the domain simulated using COMSOL simulations to determine the thermal boundary layer thickness prior to the nucleation of the bubble. (b) Thermal boundary layer thickness from the simulations for two laser powers of 50 mW and 100 mW.

Vapor bubble growth: The laser power q_{total} is dissipated by evaporation in evaporating region q_{ev} and natural convection outside the evaporating region q_{nc} (heat loss from underneath the sample is found to be negligible):

$$q_{total} = q_{ev} + q_{nc} Eq. (10)$$

Using data from Fig. 4e in the manuscript, we consider a case when $h = 120 \text{ kW/m}^2\text{K}$ and $w = 10 \mu\text{m}$, with $D_{bb} = 133 \mu\text{m}$. Our numerical simulation (Fig. 4 in manuscript) results in natural convection dissipating $\approx 55\%$ of the laser power. The thermal boundary thickness in natural convection region can be obtained by:

$$q_{nc}" = k \frac{T_{wall} - T_{bulk}}{\delta}$$
 Eq. (11)

where the area to calculate natural convection heat flux is the annulus with outer radius of 1 mm and inner radius of 140 μ m (taken from the simulation domain in Fig. 4d in manuscript), *k* is thermal conductivity of water, T_{wall} is from Fig. 4f in the manuscript, T_{bulk} is bulk liquid temperature at 25°C, and δ is thermal boundary thickness. From Eq. (11), the thermal boundary layer thickness is calculated to be $\approx 280 \ \mu$ m.

S7. Force Estimation on Complete Microlayer Wetted Bubble Base

Figure S7-a shows the main forces acting on the bubble when the entire base is wetted by the microlayer. The buoyancy force and capillary force (at the top curvature of bubble) aim to depart the bubble from the surface, while the reduced liquid pressure in microlayer (due to capillary and disjoining forces) want to keep the bubble attached on the surface. These forces are estimated below, which show that the sum of the forces that hold the bubble on the surface (1.14×10^{-5} N)

is greater than the sum of the forces that try to depart the bubble from the surface $(5.16 \times 10^{-6} \text{ N})$, thus preventing the bubble from departing even in the absence of a three-phase contact line.



Figure S6: (a) Force balance on a bubble with completely wetting microlayer, and (b) radial variation of disjoining pressure in the microlayer based on conservative estimate.

Forces aiming to depart the bubble from the surface: The buoyancy force F_b can be estimated as: $F_b = \rho_1 gV$, where ρ_1 is the displaced liquid density, g is the gravitational acceleration, and V is the vapor volume of the bubble. From Fig. 3-b2 of manuscript, we estimate the bubble volume (partial sphere geometry) to be $3.30 \times 10^{-13} \text{ m}^3$, thus resulting in $F_b = 3.23 \times 10^{-9} \text{ N}$. The pressure in liquid is lower than that inside the bubble, given by $\Delta P = \frac{2\sigma}{R}$ where σ is the surface tension of liquid-vapor interface, and R is the radius of curvature of upper liquid-vapor interface. At the top of the bubble, the bubble radius is about 35 µm (Fig. 3-b3 in manuscript). The pressure difference between the vapor and bulk liquid is 4113 Pa. This pressure is acting on a radius equivalent to the bubble base microlayer radius of 20 µm (Fig. 3-b3 in manuscript). Hence, the force due to this pressure difference is $5.16 \times 10^{-6} \text{ N}$. Thus, the sum of the forces that lift the bubble to depart it from the surface is $\sim 5.16 \times 10^{-6} \text{ N}$. Forces holding the bubble to the surface: The curvature of the microlayer is obtained from fitting a parabolic curve to the microlayer profile using data from Fig. 3-b3 in manuscript, and is estimated to be 0.02 μm⁻¹. This curvature reduces the pressure in microlayer by ~ 2880 Pa. Taking the bubble base radius of 20 μm (Fig. 3-b3 in manuscript), the force due to this pressure difference is 3.62×10^{-6} N. A conservative estimation of disjoining pressure is performed by using data for non-polar liquid due to the lack of predictive models for water. The theoretical DLVO model has many unknown parameters making its use impractical. The data of carbon tetrachloride (CCl₄) on glass is used (Carey, V. P. Liquid-Vapor Phase-Change Phenomena, 2nd ed., Taylor & Francis, 2007). Since CCl₄ is non-polar while water is polar, the disjoining pressure estimated here is lower than the real case. The disjoining pressure P_d can be determined from $P_d=A\delta^{-B}$ where where *A* is a constant of 1.782 Pa•m^B and B = 0.6 for CCl₄; δ is the film thickness. Figure S7-b shows the disjoining pressure distribution in the microlayer. The average value can be obtained from Eq. (12) as 6184 Pa, resulting in a force (7.77 × 10⁻⁶ N). Thus, the sum of the forces that hold the bubble on the surface is 1.14×10^{-5} N.

$$P_{d,ave} = \frac{\int_{1}^{20} \frac{10^4}{(0.4497 + 0.1483r)} dr}{(20 - 1)}$$
Eq. (12)

<u>Worst case Scenario</u>: In the calculation above, the microlayer thickness at the center is unknown and was assumed to be ~300 nm (which would lead to higher disjoining pressure), from which the outermost microlayer thickness was taken to be ~3 μ m. Even if we assume the maximum film thickness which allows us to see fringes as 10 μ m, the force due to the disjoining pressure will be 2.52 × 10⁻⁶ N. Thus, the sum of the forces (6.14 × 10⁻⁶ N) holding the bubble on the surface will still be greater than the sum of the forces (5.16 × 10⁻⁶ N) trying to depart the bubble from the surface. Further, P_d would be even greater for water thus further magnifying the forces holding the bubble onto the surface.

S8. Video Legends

Video 1: Steady state bubble formation on hydrophilic SiO_2 surface with degassed water. The bubble forms on the surface due to the incident laser and remains stable as the evaporation rate at the base of the bubble equals the condensation rate of vapor at the bubble's liquid-vapor interface. As the laser is blocked, the vapor within the bubble condenses causing the bubble to shrink and collapse.

Video 2: Steady state bubble formation on hydrophobic FOTS surface with degassed water due to incident laser. As expected, the bubble size is larger in size compared to the hydrophilic SiO_2 surface. The bubble achieves steady state as the evaporation rate at the base of the bubble equals the condensation rate of vapor at the bubble's liquid-vapor interface. The vapor within the bubble condenses as the laser is blocked, causing the bubble to shrink and collapse.

Video 3: Steady state bubble formation on hydrophilic SiO_2 surface with regular water containing dissolved air. The bubble size is much larger in size compared to the hydrophilic SiO_2 surface using degassed water. Further, the bubble keeps growing even at constant laser power as dissolved air is continuously released into the bubble along with evaporation of water. The vapor generation rate equals the condensation rate at the bubble's liquid-vapor interface; however, the air released into the bubble keeps accumulating causing the bubble to grow in size. As the laser is blocked, the vapor within the bubble condenses causing the bubble to shrink slightly. Now the bubble is comprised only of air and remains stable as the diffusion of air into the surrounding water (almost saturated with air) is a slow process.