Reflected laser interferometry reveals subtleties of low surface tension microdroplet condensation

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ABSTRACT

Using dual wavelength reflected laser interferometry, we developed a simple yet accurate protocol to study the dynamic behavior of condensed microdroplets. For low surface tension liquids, sufficiently sharp experimental interferograms are obtained by appropriately controlling the confocal pinhole. An automated numerical framework is developed to dynamically reconstruct the three-dimensional topography of the droplets from the obtained interferograms. The results align excellently with several theoretical trends. The framework overcomes the limitation of conventional techniques (e.g., optical imaging/environmental scanning electron microscopy) and allows quantitative investigation of the condensation of low-surface-tension liquids under atmospheric conditions. We also report a spontaneous thermocapillary motion during condensation where small microdroplets moved to relatively larger droplets in the vicinity. This movement is exclusive to a combination of low surface tension liquids and hydrophilic surfaces. We attribute this motion to two factors, namely, formation of an ultrathin precursor film on hydrophilic surfaces and a gradient in local temperature inside a small microdroplet caused by the difference in adjacent droplet sizes. Although we have demonstrated the framework for dropwise condensation, the protocol finds direct applicability in the study of a wide range of fundamental phenomena involving droplet dynamics which includes wetting, spreading, droplet pinning and motion, contact angle hysteresis, droplet coalescence etc., and suitable adaptation of the developed protocol can assist in unveiling previously unknown fundamental physical processes of practical relevance.

1. Introduction

Microdroplet condensation has attracted sustained attention because of its various engineering applications, such as water harvesting [1] and condensation heat transfer [2]. Studies involving condensed microdroplets with high contact angles progressed using environmental scanning electron microscopy (ESEM) because of good spatiotemporal resolution [2-4]. However, the vapor available for droplet condensation in ESEM chamber is restricted to water, which provides high contact angles due to high surface tension. The observation of the condensation of low surface tension liquid, such as alcohol and refrigerant, has been normally conducted by optical microscopy [5], although it is difficult to precisely measure the contact angles of the microdroplets due to low spatial resolution and depth of focus. Consequently, there exists major gaps in our understanding of the condensation behavior of microdroplets with low contact angles.

Reflection interference contrast microscopy (RICM) is an alternative and promising way to observe the dynamics of microdroplet condensation. RICM can provide interference patterns corresponding to local droplet thickness, which are essential for understanding the dynamic behavior of condensed microdroplets. In addition, RICM enables us to precisely observe microdroplet condensation of low surface tension liquids because there is no restriction of liquids. However, such studies using RICM are very rare [6], and consequently the scientific endeavor on the characterization of microdroplet condensation of low surface tension fluids remains relatively scarce.

In our work, we used dual-wavelength reflection interference contrast microscopy (DW-RICM) in confocal mode to develop a framework to capture and quantify the dynamics of droplet condensation and demonstrate the capability of our framework by studying the dynamics of condensation of acetone and ethanol, test liquids with low surface tension and high vapor pressure, on surfaces with different surface energies. Sharp interferograms are obtained by controlling the confocal pinhole of the RICM imaging setup. A novel image processing framework [7] has also been developed to interpret the experimental interferograms. Utilizing the reflected intensity pattern corresponding to two monochromatic laser sources, the drop shape is reconstructed unambiguously. We have performed a complete 3D reconstruction of the drop shape as well which allows us to extract several other valuable quantitative data, namely, evolution of drop volume, liquid–vapor surface area and footprint of the droplets. It is to be noted here that the developed interferometric protocol enables studies on droplet condensation at precisely controlled pressure, humidity, and temperature, which is not achievable in environmental ESEM. Using the framework, we also discovered [8] a novel directional motion of low surface tension microdroplets when they nucleate on high surface energy surfaces.

Experimental Details

Microdroplet condensation experiments are conducted in a in a flow cell made of UV quartz (49UV2 - Micro Demountable Flow Through Cell, Fireflysci Inc., Staten Island, NY). First, the liquid cell (with the lid in place) was partially filed with 5 ml of test liquid. Then 2-3 ml of air was infused into the liquid cell, which forms an air bubble. The liquid cell was slightly tilted thereafter allowing the air bubble to move up to the centre of the liquid cell (the observation area of the microscope) due to buoyancy. Then the two inlets of the liquid cell were covered with cover glass to prevent evaporation of the test liquid. A schematic representation (not to scale) of the experimental set up is presented in Figure 1(a). After putting the experimental set up on the observation stage of DW-RICM, a drop of cold water is placed on the outer surface of the lid of the liquid cell on top of the observation area to locally reduce the temperature. It helps in promoting condensation of the vapor of the high vapor pressure test liquid present inside the liquid cell.

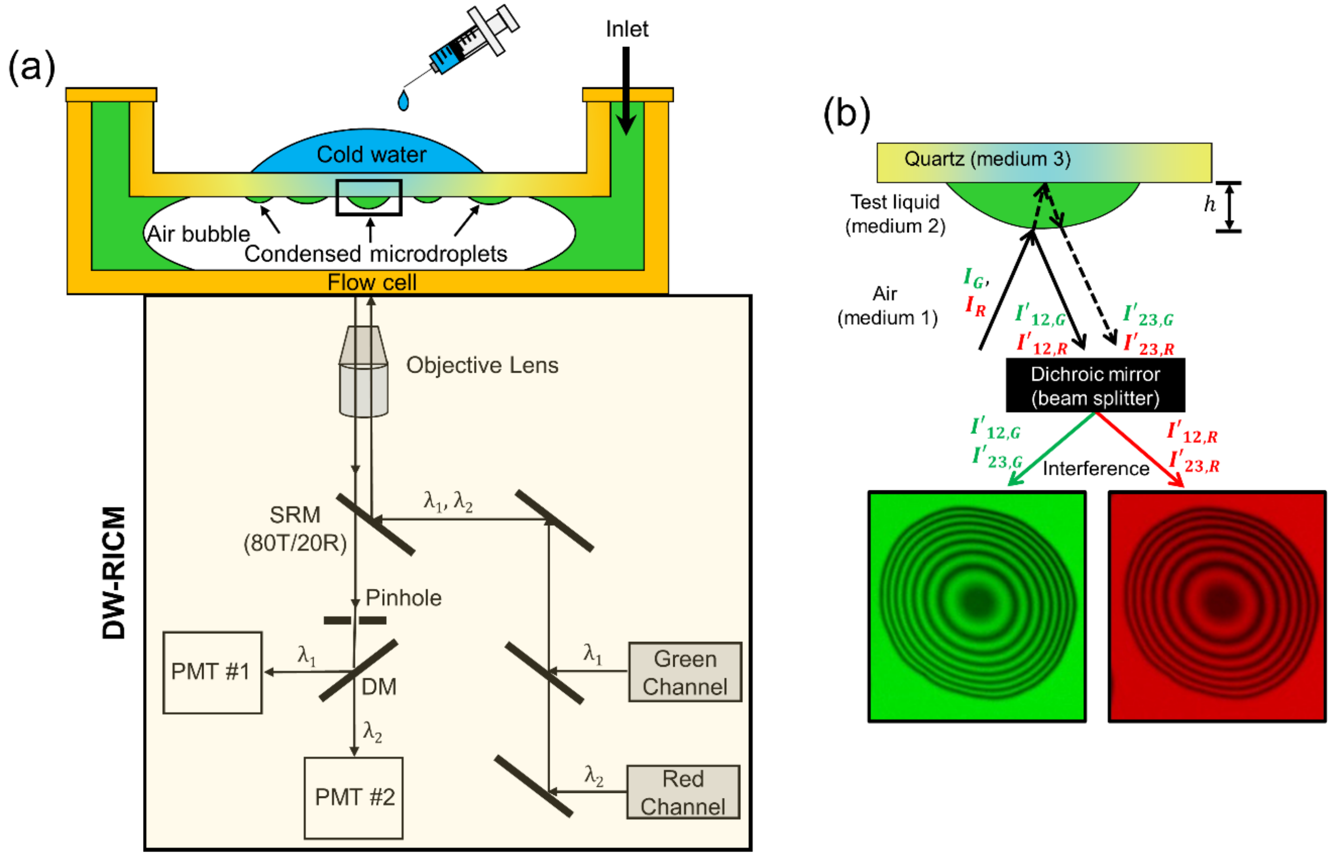

Figure 1: (a) Schematic representation (not to scale) of the experimental setup for droplet condensation, (b) Simplified light path of the DW-RICM setup enabling interferometric imaging of the condensation dynamics from bottom view.

The condensation dynamics is captured with LSM 800 (Carl Zeiss, Canada), a confocal dual wavelength reflection interference contrast microscope (DW- RICM), using a 10X objective lens with numerical aperture of 0.3 (EC Plan-Neofluar, Carl Zeiss, Canada). The observation area was scanned pixel-by-pixel simultaneously using two focused beams of monochromatic laser with wavelengths 488 and 561 nm and the reflected light was collected through the pinhole of the confocal microscope before being routed to two photomultiplier tubes, each corresponding to one of the laser wavelengths. A simplified light path of the setup is presented in Figure 1(b). The detected analog optical signal is then converted to a digital image using an inbuilt deconvolution algorithm. The pinhole size was kept at 1 airy unit. For the used objective lens, it corresponds to an in-focus axial slice length of ~ 11 µm when operating in dual wavelength mode. This is sufficient for capturing the entire 3D topography of low surface tension microdroplets with maximum height ~ 4-5 µm. At this pinhole size, we were able to get sufficiently strong signal from region of interest, while avoiding stray light (noise) from out of focus planes. The pixel size and scan area were chosen such that the dwell time at each pixel was less than 1 µs, which is necessary to have satisfactory temporal resolution for capturing the dynamic behavior.

In the DW-RICM setup, the incident light rays get reflected from two interfaces, namely quartz/test liquid and test liquid/air interfaces, interfere with one another (see Figure 1(b)). This interference results in the occurrence of bright and dark fringes depending on the difference in the light path. Difference in local height of the droplet between adjacent bright and dark fringes corresponds to $\lambda/(4n)$, where λ is the wavelength of the used laser source and n is the refractive index of the droplet. Based on these fundamentals, we developed a holistic framework [7] (details not included here) to perform complete topological reconstruction of condensing microdroplets with a vertical resolution of approximately 90 nm. The analytical framework also enables us to obtain quantitative data regarding the growth and motion of the droplets (e.g., contact angle, height, surface area, volume, velocity of motion etc.).

Results and Discussion

Studying growth and coalescence of condensed acetone microdroplets

Using the DW-RICM framework, we experimentally captured the growth of acetone microdroplets on the quartz surface. During its lifecycle, a condensing droplet undergoes two different types of events, namely, growth events – when the droplet grows uninterruptedly due to condensation without encountering nearby droplets and coalescence events – when, as the droplet grows, its contact line touches other neighboring droplets, and they merge to form a bigger droplet. In Figure 2, the droplet of interest (denoted by 1) underwent four such growth events (namely, G1, G2, G3 and G4) and three coalescence events (namely, C1, C2 and C3) during a total observation period of 75.85 s. A total of 185 frames for each of the two wavelengths were captured over this observation window, which provided us with sufficient temporal resolution to capture the dynamics of the process.

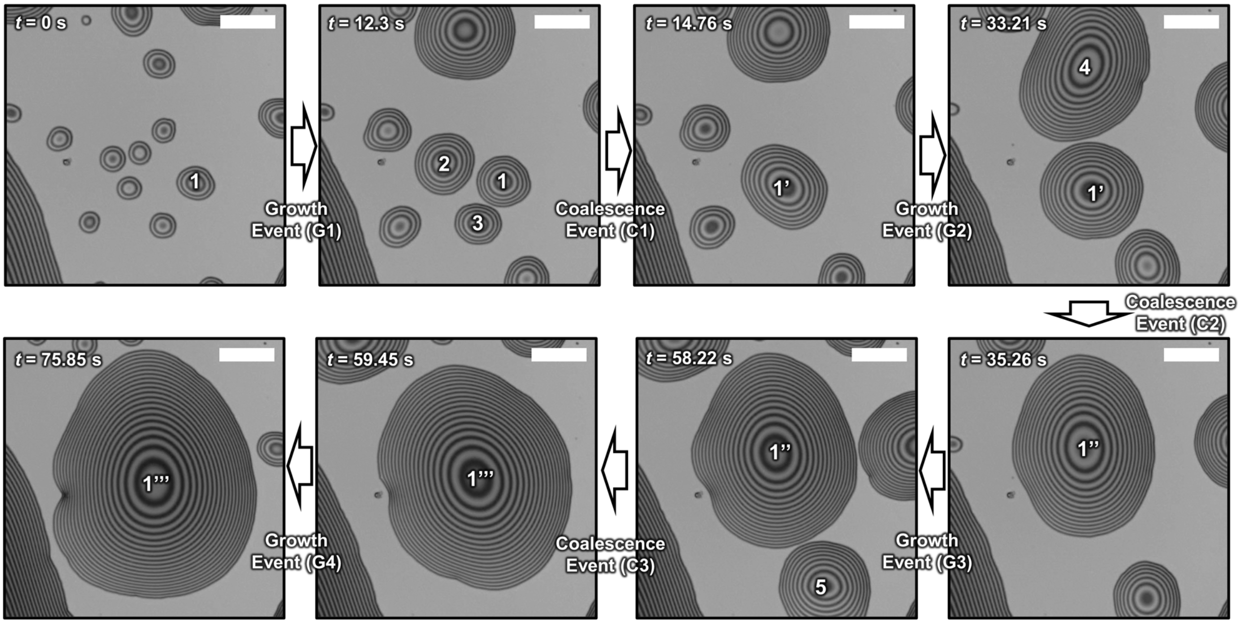

Figure 2: Time evolution of a typical acetone microdroplet condensation event.

Using our in-house analytical framework, we also analyzed the growth of the microdroplet denoted by 1 in Figure 2. The results are presented in Figure 3.

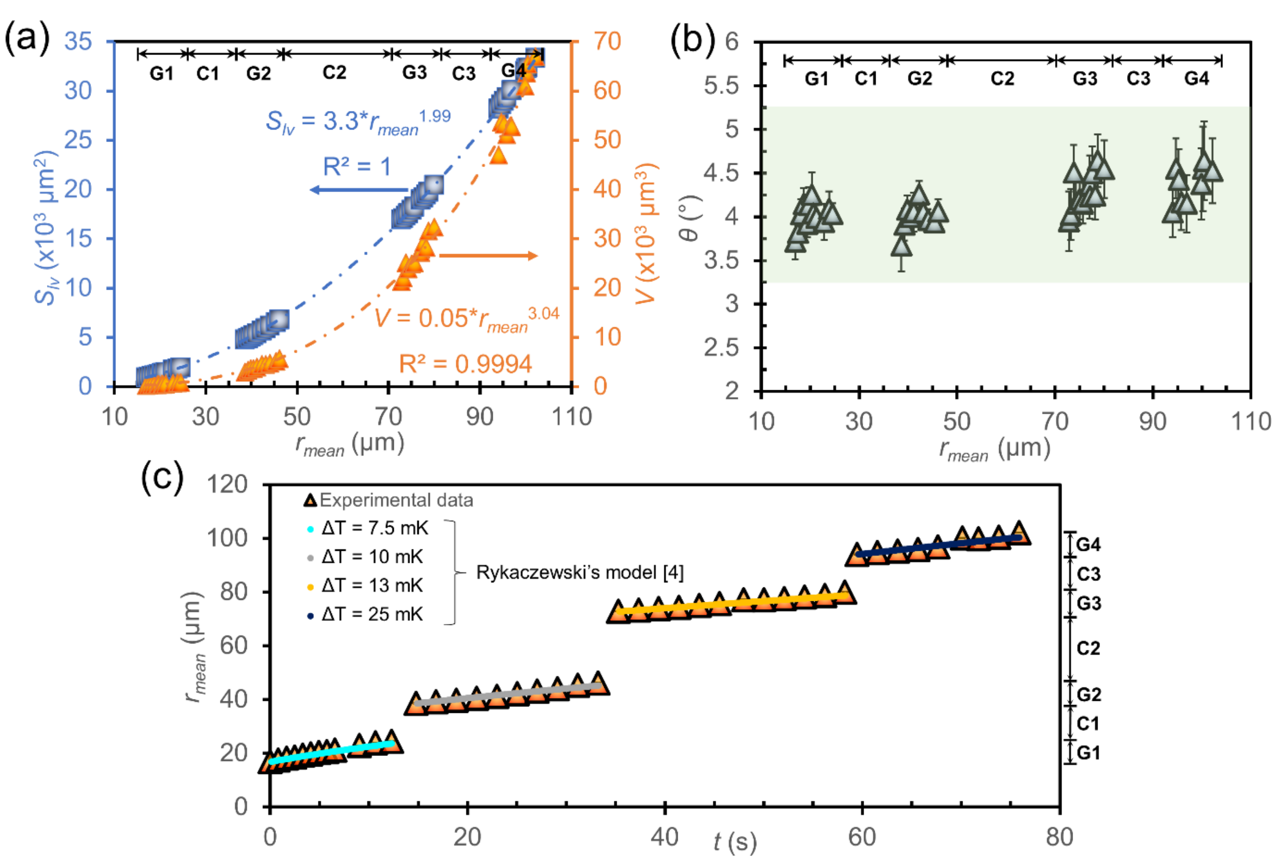

Figure 3: Quantitative analysis of the dynamics of growth of a condensing acetone microdroplet. (a) Variation of droplet volume V and liquid–vapor surface area S_{lv} with mean base radius. (b) Dependence of the mean contact angle θ on mean base radius. (c) The evolution of mean base radius r_{mean} with time during condensation.

The measured values of volume, V, and the liquid–vapor surface area, S_{lv} are plotted for different mean base radii, r_{mean} in Figure 3(a). By fitting a power law profile to the experimentally determined dependences $S_{lv}(r_{mean}), V(r_{mean})$, we obtained $S_{lv}{\sim}r_{mean}{}^{1.99}$ and $V{\sim}r_{mean}{}^{3.04}$, which stand in excellent agreement with the analytical formulas for an ideal spherical cap [9] with a constant contact angle where $S_{lv}{\sim}r_{mean}{}^{2}$ and $V{\sim}r_{mean}{}^{3}$. Therefore, the shape of the condensed microdroplets during condensation and growth phase can be approximated by the spherical cap shape. Also, the contact angle remained almost constant (within 3.25º–5.25º) and the values were effectively invariant with respect to changes of the base radii, which is an expected outcome resulting from the combination of a low surface tension condensate and a near smooth surface with a measured arithmetic mean surface roughness as low as 2.9 nm.

Additionally, we have also captured the growth of the mean base radius r_{mean} with time in Figure 3(c) and compared it to the theoretical prediction of Rykaczewski [4] for droplet growth in constant contact angle mode. As can be seen in Figure 3(c), the experimental data could be excellently fitted to the model of Rykaczewski [4] with ΔT being the fitting parameter. Note that ΔT was assumed to be constant during each of the pure growth phases as a simplification and updated after each coalescence event. After each coalescence event, the fit yielded a higher ΔT value for the next growth phase, as the drop height underwent a noticeable increase following coalescence and consequently the heat transfer resistance between the saturated vapor and the base surface increased**.**

Unveiling novel motion of low surface tension microdroplets on high surface energy surfaces:

Using our experimental framework, we also revealed [8] a spontaneous motion of low surface tension microdroplets on high energy surfaces (e.g., quartz) which was induced by a thermal Marangoni effect and an ultrathin precursor film. Figure 4 shows a condensation event on a quartz surface involving ethanol as the low surface tension test liquid. The entire observation area is shown in Figure 4(a). Several small microdroplets were nucleated in the central region of the observation area. In addition, two large droplets were in the left and right sides of the observation area. Figure 4(b) shows a typical 3D profile of an ethanol microdroplet reconstructed based on our analytical framework. The contact angles of the observed microdroplets were constant at approximately 5° and are independent on the base radius. As shown in Figure 4(c), temporal evolution of a microdroplet (indicated by a white arrow) was tracked from its nucleation. The droplet nucleated and got pinned at an impurity indicated by a black arrow. During its growth, the center of the droplet clearly shifted to the left side. At 81.6 seconds, the three-phase contact line was eventually depinned from the impurity, as indicated by the red arrow. After the depinning, the microdroplet immediately moved to the left side and then merged with a bigger droplet. This indicates that a pulling force is acting on the droplet which makes it move towards the left side. In addition, the microdroplets indicated by red arrows in Figure 4(a) moved to the big droplet in the left side, while the microdroplets denoted by yellow arrows approached the big droplet in the right side. This result indicates that the smaller droplets always move towards a nearby bigger droplet.

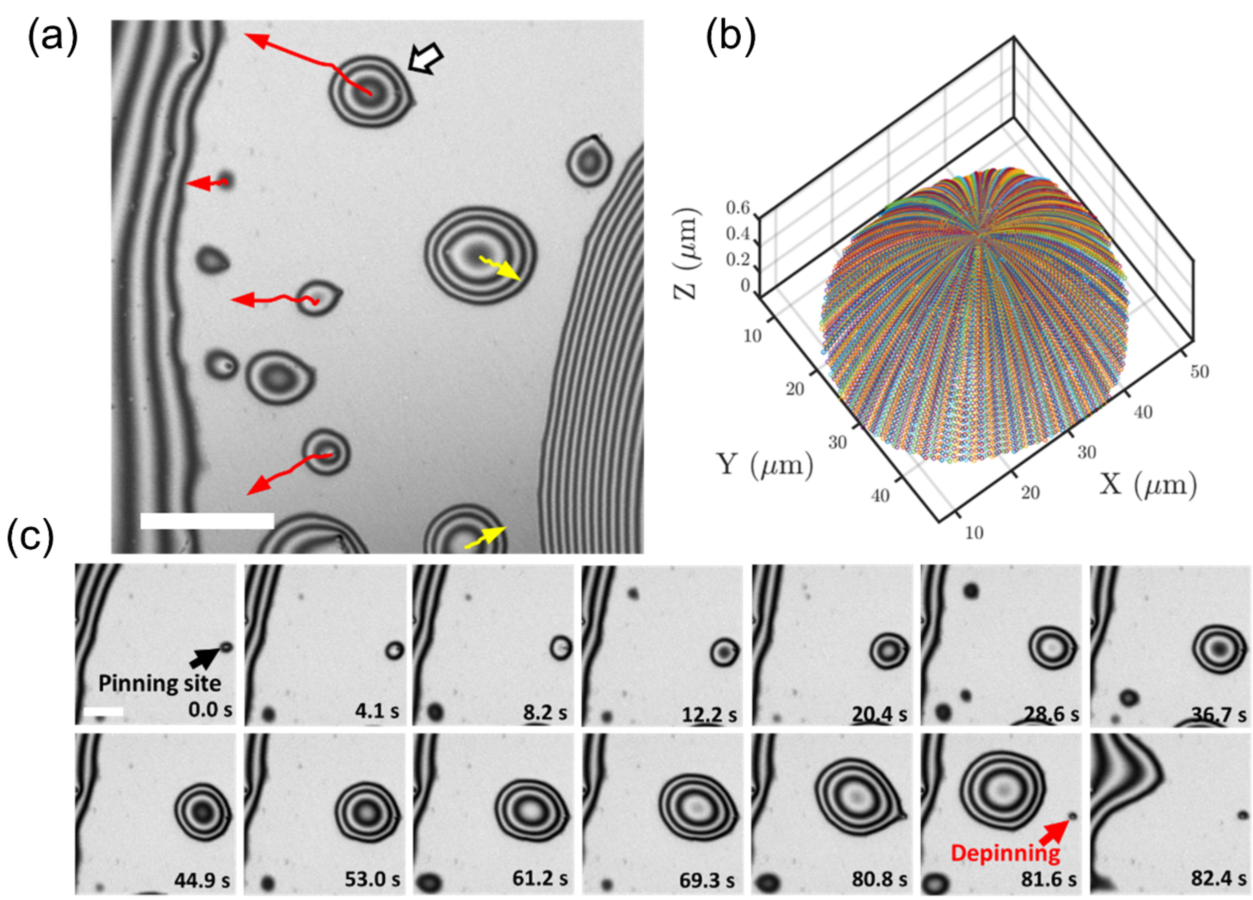

Figure 4: (a) Interferometric image showing the experimental domain for ethanol microdroplet condensation on quartz surface. Red and yellow arrows track the center position of the nucleated droplets. (b) 3D profile of an ethanol microdroplet reconstructed from its interference patterns. (c)

Timeseries demonstrating the nucleation, growth, and movement of a microdroplet indicated by a white arrow in (a). Scale bars in (a) and (b) show 50 μm and 20 μm, respectively.

We also conducted water condensation on the same quartz surface and ethanol condensation on a hydrophobic polydimethylsiloxane surface (data not shown), but the droplet movement was not observed in both the cases. This is an unambiguous indication that this motion is realizable exclusively for a combination of low surface tension liquid and hydrophilic solid surface.
During the movement (at 81.6 seconds in Figure 4(c)), the droplet showed a contact angle hysteresis of 0.5º, which is much lower than the ones typically observed for macroscopic droplets (20-50º) [10] and is within the typical hysteresis range observed on slippery lubricant-infused porous (SLIPS) surfaces (<3º) [10]. From these results, we conclude that vapor of low surface tension liquid adheres to high energy surface and forms an ultrathin liquid film (precursor film) during condensation. The precursor film works as a lubricant layer and thus decreases the resistance for droplet movement, in the same manner as a lubricant on SLIPS surfaces.

Conclusion:

Using dual wavelength reflected laser interferometry, we developed a simple yet accurate protocol to study the dynamic behavior of condensed microdroplets. For low surface tension liquids, sufficiently sharp experimental interferograms are obtained by appropriately controlling the confocal pinhole. An automated numerical framework is developed to dynamically reconstruct the three-dimensional topography of the droplets from the obtained interferograms. We also report a spontaneous motion during condensation where small microdroplets moved to relatively larger droplets in the vicinity. This movement is exclusive to a combination of low surface tension liquids and hydrophilic surfaces. Although we have demonstrated the framework for dropwise condensation, the protocol finds direct applicability in the study of a wide range of fundamental phenomena involving droplet dynamics which includes wetting, spreading, droplet pinning and motion, contact angle hysteresis, droplet coalescence etc., and suitable adaptation of the developed protocol can assist in unveiling previously unknown fundamental physical processes of practical relevance.

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