Role of entrapped vapor bubbles during microdroplet evaporation

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On superheated surfaces, the air bubble trapped during impingement grows into a larger vapor bubble and oscillates at the frequency predicted for thermally induced capillary waves. In some cases, the entrapped vapor bubble penetrates the droplet interface, leaving a micron-sized coffee-ring pattern of pure fluid. Vapor bubble entrapment, however, does not influence the evaporation rate. This is also true on laser heated surfaces, where a laser can thermally excite capillary waves and induce bubble oscillations over a broad range of frequencies, suggesting that exciting perturbations in a pinned droplets interface is not an effective avenue for enhancing evaporative heat transfer. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4745009]

Cavities of entrapped gas (i.e., bubbles) in working fluids and at fluid interfaces play an important role in the functionality of many systems. In some cases, the presence of bubbles can be advantageous. For example, in heat transfer applications (e.g., spray cooling and boiling), entrapped air bubbles at the liquid-solid interface act as vapor nuclei and initiate nucleate boiling at lower superheats than theory predicts.1 In fuel combustion processes, entrapped vapor bubbles near the liquid-gas interface can activate a self-ignition mechanism based on vapor bubble cavitation.2 Trapped air cavities in direct-write applications promote the formation of non-wetting droplets on structured surfaces.3 On the other hand, air bubbles also hamper system reliability by perturbing the pressure field inside the printhead.4 Likewise, in lab-on-chip systems, nano- and micro-bubbles can both promote and disrupt system performance by altering the pressure and temperature profiles in the fluid channels.5 For all these examples, it would be ideal to predict a priori the dynamics of the bubble interface responding to fluctuations in system temperature and pressure. In the case of thermal fluctuations, the roughness of a liquid-vapor interface can be described in terms of thermally excited capillary waves. Thermally excited capillary waves are essential for predicting related effects like the spontaneous rupture of thin liquid films and droplet coalescence.6,7

This work focuses on the correlation between thermally excited capillary waves and the evaporation rate of water microdroplets that retain entrapped vapor bubbles. When a microdroplet impacts a surface, a small air bubble can be trapped at the droplet-surface interface. On isothermal surfaces heated above the saturation temperature, this trapped air bubble grows into a larger bubble containing air and water vapor. It then undergoes a complex, oscillatory interplay between evaporative growth and condensation. This work shows that the frequency of these oscillations correlates well with those predicted for thermally induced capillary waves on a free surface. In addition, experiments are conducted with water microdroplets on laser heated surfaces as a function of laser heating frequency. For laser heated surfaces, the microdroplets profile (i.e., apex height and contact angle) oscillates at the heating frequency. Yet, the evaporation rate is nearly constant (aside from a slight increase at heating frequencies within 500 kHz to 10 MHz). The remainder of this letter describes these results.

The experiments were performed using a standard imaging/dispensing apparatus. See Fig. 1 in the supplemental material.8 The setup consists of a microdroplet ink-jet dispenser, a horizontal sample stage (with temperature control), high-speed visible and infrared (IR) cameras, and a femtosecond pulsed laser system equipped with an electro-optic modulator. Experiments comprised of dispensing single microdroplets of deionized water on an Al thin-film having either (1) a controlled (isothermal) or (2) a time-varying (modulated) surface temperature. For the isothermal studies, the surface temperature is regulated by a temperature controller coupled to a heating/dispensing apparatus. See Fig. 1 in the supplemental material.8

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Role of entrapped vapor bubbles on isothermal surfaces: Previous studies by the authors explored the correlation...
between the evaporation rate and parameters such as the surface temperature, contact line dynamics, and sample surface energy and/or structure.\textsuperscript{9,10} There discussion on the dynamics of air-vapor bubbles trapped inside the evaporating microdroplets was omitted. As mentioned previously, air bubble entrapment can occur during the impingement process and is particularly important in heat transfer because the air bubbles can act as vapor nuclei and initiate nucleate boiling at much lower superheats than theory predicts.\textsuperscript{1} When air bubbles are trapped on isothermal surfaces heated below the saturation temperature, the air bubble dynamics are monotonous, remaining fixed in size and position with infrequent detachment from the surface. On superheated surfaces, however, the trapped vapor bubble dynamics are complex, undergoing vapor nucleation, growth, condensation, and oscillations. In many experiments on superheated surfaces, one or more vapor bubbles will nucleate after impact. Typically, this requires surface temperatures above \( \sim 110^\circ C \). In this case, the vapor bubble nucleation sites are not correlated with any specific surface location under the microdroplet (aside from defects in the Al surface layer that are not resolved by the camera in these experiments).

Fig. 1(a) shows a series of high-speed images for an evaporation experiment at \( T_S \approx 189^\circ C \) where two vapor bubbles nucleated after impingement on an Al thin-film. The vapor bubbles combined into one bubble at \( t \approx 17.3 \) ms. A subsequent image at \( t \approx 17.5 \) ms is displayed. After combination, the vapor bubble underwent a complex interplay between evaporative growth and condensation, coinciding with steady-state vapor bubble oscillations at kHz frequencies. In this case, the frequency of these steady-state vapor bubble oscillations is in good agreement with the frequency for thermally excited capillary waves. For example, the measured vapor bubble oscillations are within 23.3 kHz \( \leq \omega_{VB} \leq 24.3 \) kHz, while the calculated thermally excited capillary oscillations are \( \sim 17 \) kHz. The capillary frequency is calculated using the relation\textsuperscript{11}

\[
\omega_0 = \left( \frac{\sigma k^3}{\rho} \right)^{1/2},
\]

where \( \sigma \) and \( \rho \) are the surface tension and density of water, and \( k \) is the capillary wavenumber. Fig. 1(b) compares experimental data with the dispersion behavior predicted by Eq. (1). For this comparison, the wavenumber is approximated as \( k = 1/D_v \), where \( D_v \) corresponds to the measured contact diameter of either the pinned vapor bubble (x = VB) or water microdroplet (x = droplet). Steady-state oscillations were only observed at surface temperatures within 175 °C \( \leq T_S \leq 215^\circ C \). At lower superheats, the oscillations are unsteady. At surface temperatures beyond \( \sim 215^\circ C \), the Leidenfrost effect comes into play, resulting in exploding vapor bubbles and bouncing microdroplets (well before oscillations can persist). A unique result of a vapor bubble penetrating the water-vapor interface is also shown in Fig. 1(a). In this case, the trapped vapor bubble penetrates the microdroplet interface and forms a micron-sized “coffee-ring” pattern. Thus, vapor bubble entrapment can serve as another physical mechanism to facilitate coffee-ring evaporation patterns.\textsuperscript{2} These coffee-ring patterns of pure water only formed on Al thin-films superheated within 175 °C \( \leq T_S \leq 190^\circ C \).

Role of entrapped vapor bubbles on laser heated surfaces: These experiments were performed with recognition that instabilities at liquid interfaces could play an important role during evaporation. Laser heating experiments covered a broad range of heating frequencies, perturbing the microdroplet at frequencies that coincide with specific surface capillary waves. Fig. 2 shows the evaporation results as a function of laser heating frequency at constant duty cycle (50%), heating power \( (P_{laser} = 144 \pm 2 \) mW), and laser beam waist \( (w_{laser} = 33.6 \) \( \mu \)m). This data do not discriminate between experiments with or without vapor bubble nucleation. For example, vapor bubbles nucleated in slightly over half the experiments conducted—at some frequencies slightly more than others—and all the acquired data was analyzed regardless of bubble nucleation or not. Thus, the presence of vapor bubbles did not change the evaporation rate beyond the variation provided in Fig. 2. High-speed video measurements were capable of resolving vapor bubble
oscillations for heating frequencies up to $f_{\text{mod}} \approx 100$ kHz. The amplitude of these oscillations scaled inversely with the heating frequency. In fact, for $f_{\text{mod}} \lesssim 55$ kHz, the vapor bubble and droplet profile (i.e., $\theta$ and $h$) oscillated at the heating frequency (i.e., not at $f_{\text{mod}}/2$). Higher-speed data acquisition and improved image resolution are required for $f_{\text{mod}} \gtrsim 100$ kHz. See supplemental material.\(^8\) In experiments when vapor bubbles did not nucleate at the laser heating center, oscillations in the droplet profile were less than measurement resolution ($\pm 2\mu m$), especially at $f_{\text{mod}} \approx 1$ kHz. Nevertheless, as shown in Fig. 2, such oscillations did not change the evaporation rate beyond the variation provided.

Fig. 2 provides experiment data at different relative humidity ($RH$) conditions of the laboratory. The $RH \approx 50\%$ data (triangles) was acquired after analysis of the $RH \approx 5\%$ data (circles) to both confirm the results and add additional data points at $f_{\text{mod}} \approx 2.7$ kHz and $f_{\text{mod}} \approx 2.7$ MHz. First glance at Fig. 2(a) suggests that the relative humidity has a slight influence on the evaporation rate. However, the increase in humidity coincided with slight changes in the droplet wetting behavior. The average contact radius and contact angle for both data sets are provided in the figure caption. To account for variations in contact radii, Fig. 2(b) provides the evaporation rate per unit length of contact line. As shown in Fig. 2(b), the relative humidity of the room has no effect on the evaporation rate per unit length of contact line. This result correlates with our previous studies on iso-thermal surfaces, where the evaporation rate per unit length of contact line is insensitive to moderate changes in sample surface energy (i.e., wetting behavior).\(^9\)

There may be a slight enhancement in the evaporation rate at heating frequencies within the range $500$ kHz $\lesssim f_{\text{mod}} \lesssim 10$ MHz. Although this increase is small and arguably within the experimental variation provided, for comparison, this heating frequency range is compared with $\omega_l$ (i.e., Eq. (1)). In this case, $500$ kHz $\lesssim f_{\text{mod}} \lesssim 10$ MHz corresponds to $1600$ cm$^{-1} \lesssim k \lesssim 11700$ cm$^{-1}$ and thermal penetration depths ($\ell_T \propto 1/k$) within $6.3 \mu m \gtrsim \ell_T \gtrsim 0.85 \mu m$. Thus, if instabilities in the viscoelastic properties of interfacial water play a role in evaporation, then such instabilities are most important at length-scales within the range of $1 \mu m \gtrsim \ell_T \gtrsim 7 \mu m$. Another way to interpret these results is to consider the correlation between the evaporation rate and changes in the droplet’s interfacial surface area. In this case, the amplitude of these surface capillary waves may be maximized at specific vibrational modes. In result, high frequency capillary waves may slightly increase the evaporation rate by effectively increasing the roughness (i.e., liquid-vapor interfacial area). This, of course, is only valid if surface ripple and curvature effects do not influence the accommodation coefficient. High frequency capillary waves on millimeter-sized water droplets at MHz frequencies have been observed by others,\(^13\) giving us some confidence in this interpretation. Interestingly, the slight increase in the evaporation rate at MHz heating frequencies correlates well with the heating frequency used to heat water in a microwave oven (i.e., 2.45 MHz).

In summary, the dynamics of entrapped vapor bubbles may be predicted in terms of thermally excited capillary waves. In some cases, the existence of a pinned vapor bubble inside an evaporating microdroplet will facilitate the formation of a liquid coffee-ring structure. Vertical displacements of a droplets liquid-vapor interface can be achieved by simply modulating the surface temperature. However, the effects of changing the surface heating frequency on the evaporation rate are small, suggesting that exciting perturbations in the microdroplet’s shape is not an effective avenue for enhancing evaporative heat transfer.

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