Levitation and Self-Organization of Liquid Microdroplets over Dry Heated Substrates

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Levitating droplets of liquid condensate are known to organize themselves into ordered arrays over hot liquid-gas interfaces. We report experimental observation of similar behavior over a dry heated solid surface. Even though the lifetime of the array is shorter in this case, its geometric characteristics are remarkably similar to the case of droplets levitating over liquid-gas interfaces. A simple model is developed that predicts the mechanisms of both droplet levitation and interdroplet interaction leading to pattern formation over a dry surface; the model is shown to be in good agreement with the experimental data. Using the insights from the new experiments, we are able to resolve some long-standing controversies pertaining to the mechanism of levitation of droplets over liquid-gas interfaces.

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The ability of microscale droplets of condensed liquid to levitate over a hot liquid-gas interface has been known for many decades and is thought to be the cause of white mist sometimes seen over a surface of hot tea or coffee [1]. However, careful experimental studies of this phenomenon have been conducted only recently in the works of Fedorets *et al.* [2–4] and Umeki *et al.* [5]. They observed a remarkable degree of order and size uniformity among levitating droplets. The highly localized heating in the setup developed by Fedorets [2] resulted in localization of the droplet array, leading to the introduction of the term "droplet cluster," while Umeki *et al.* [5] mostly focused on collectively moving arrays of levitating droplets. A typical photograph of such an array formed over a surface of water is shown in Fig. 1 (top). From the practical standpoint, these experimental discoveries show that our understanding of heat and mass transfer processes occurring near liquid-gas interfaces is in fact incomplete since formation of arrays of levitating droplets can have a significant effect on the heat transfer rates, especially on microscale and in the regions of intense evaporation. Furthermore, understanding the criteria for levitation or repulsion versus impact of microdroplets near interfaces is important for applications in spray cooling [6], drug delivery by means of aerosol inhalation [7], and synthesis of amorphous chemical components via containerless processes [8].

Recent studies aimed at better understanding of the physics of levitating droplets over hot liquid surfaces resulted in many suggestions of possible mechanisms of levitation and self-organization including electrostatic interactions, thermocapillary forces, liquid-liquid Leidenfrost effect, electrical charges in the gas phase, and Stokes drag force from the upward flow of air-vapor mixture [2–5,9–11]. However, the mechanisms of levitation and ordering remain matters of controversy, as none of the theoretical models is able to quantitatively predict the key observations reported by Fedorets *et al.* [2–4] and Umeki *et al.* [5], most notably the height of droplet levitation and interdroplet spacing as functions of the droplet size. We developed an experimental setup which allowed us to observe regular arrays of levitating microdroplets not only over the liquid surface but also over *dry* heated solid substrate. We note that while droplet levitation over dry

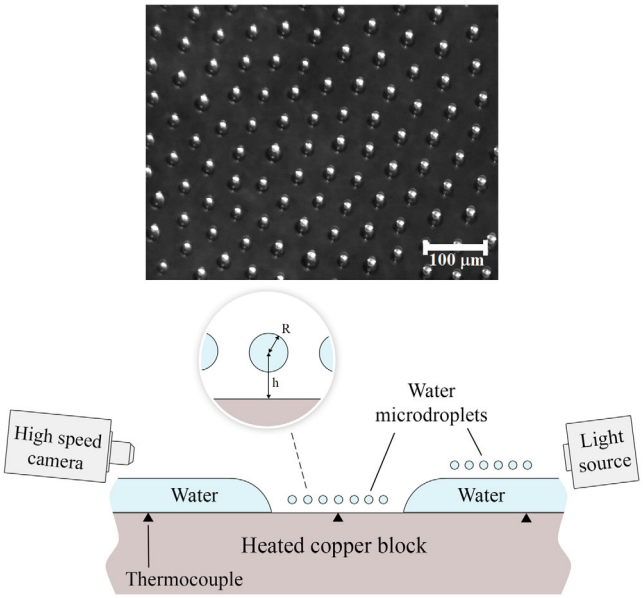

FIG. 1. (top) Array of levitating droplets over the liquid-gas interface viewed from the top. (bottom) Sketch of the experimental setup used for studies of droplet arrays over both liquid surface and dry substrate; droplet radius R and levitation height h are shown.

heated surfaces has been observed in the numerous experimental studies of Leidenfrost phenomena [12–14], self-organization of droplets into regular structures has not been seen in these configurations. Furthermore, the well-developed models of levitation of Leidenfrost droplets are not applicable to the present situation since the substrate temperatures in our experiments are far below the Leidenfrost temperature. Physical effects routinely neglected in these models, such as diffusion and Stefan flow (gas flow compensating for the diffusion of air molecules toward the interface) become important at these relatively low temperatures; all of these are taken into account in our modeling approach presented below.

The sketch of the experimental setup is shown in Fig. 1 (bottom). The substrate is a copper block heated from below. In order to achieve pinning of the contact line at the substrate, the working surface was rough with the root mean square (rms) roughness of 0.50 μm, based on measurements by an atomic force microscope. The surface temperature of the block T_w is measured by thermocouples at several points, confirming that the condition $T_w = \text{const}$ is satisfied along the copper surface. Degassed ultrapure water (Merck Millipore) is used as the working liquid. The wettability of the working surface was measured at different points using a drop shape analysis system (DSA100 by Krüss GmbH). At room temperature, the static advancing contact angle is $77 \pm 6°$, whereas the static receding contact angle is $15 \pm 4°$. An optical recording is made at 5400 frames per second using a high-speed CCD camera equipped with a microscope objective of high resolving power. The field of view of the camera is 800×800 μm^2 (spatial resolution of 0.781 μm per pixel, leading to relative error in the determination of droplet sizes between 2% and 13%). The camera is oriented either for top view or for side view. In the latter case, the optical axis of the camera is aligned at an angle of 5° with respect to the horizon. The ambient temperature is 25 °C, humidity is about 60%. Images from the camera are processed using SPIP software (Image Metrology). In experiment, the working liquid is deposited with a syringe onto the substrate to form a liquid layer of the initial thickness of 0.40 mm. With a short pulse of air jet a dry spot (about 0.5–1 mm in size) is formed on the copper surface. The heater is then switched on, resulting in evaporation and formation of droplet array, seen moving over the liquid surface in a collective fashion.

We started by observation of droplets over the liquid-air interface, with all dynamics consistent with previous experimental observations [2,5]; the photo shown in Fig. 1 (top) is a result of one of these runs at $T_w = 85$ °C (see also videos S1 and S2 in the Supplemental Material [15]). Experimental observations show that as the array approaches the contact line, see Fig. 2 (top), it encounters strong upward flow from the region of intense evaporation near the contact line. This flow results in the breakup of ordering, with some droplets being carried away from the interface and some flying over the contact line and ending up over the dry surface of the heater. Remarkably, however, the impact of such droplets on the dry surface is almost never observed. Instead, they end up levitating over the solid while shifting towards the contact line and evaporating over a time span of a few seconds. Droplets that evaporate to some critical size end up rapidly rising away from the wall. Note that all these phenomena are happening at the temperature well below the Leidenfrost temperature of water and even below the saturation temperature; the levitation distance is on the order of droplet size or higher. Most importantly, the top view in Fig. 2 (bottom left) clearly shows that the droplets arrange themselves into an array with interdroplet separation being of the same order of magnitude as the array spacing for levitation over the liquid surface, although not with the perfect regularity seen in Fig. 1, in part due to continuous bombardment of the array by droplets arriving from the contact line region. Self-organization of droplets over a *dry* surface rather than liquid-vapor interface has never been observed before.

Let us now discuss possible physical mechanisms of the observed droplet levitation over heated solid substrates. One is the Marangoni stress that can generate the flow into the space between the droplet and a flat interface [16]. However, this is not possible in our configuration since the bottom of the droplet should be at a lower temperature than the top for that mechanism to be operational. This is highly unlikely given that the only source of heat in the system is the heated metal solid substrate which is essentially at constant temperature. The second possible mechanism is

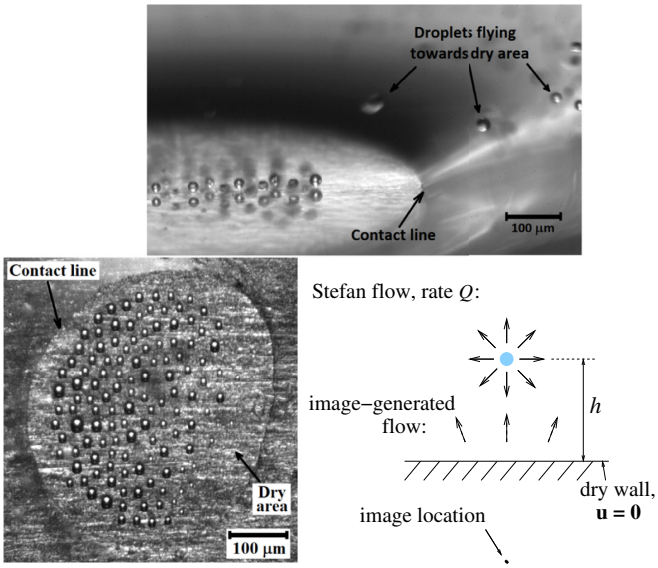

FIG. 2. Photos of levitating water droplets: (top) Side view of droplet fly-over near the contact line at $T_w = 90$ °C; (bottom left) array of droplets over the dry patch viewed from the top at $T_w = 85$ °C; (bottom right) sketch for the derivation of levitation condition. See also videos S3 and S4 in the Supplemental Material [15].

thermophoresis. We estimated the local temperature gradient near the heated substrate, based on temperature measurements by thermocouples, and substituted it into the formula for the thermophoretic force in the limit of large Knudsen numbers [17]. The result is orders of magnitude below the force needed for levitation. The electrical charges possibly carried by droplets cannot cause levitation since droplet interaction with its electrostatic image is attractive (the image charge is of the opposite sign since the substrate is a metal plate).

In an effort to explain the levitation phenomena, we now focus our attention on the effect of evaporation. For simplicity, let us assume that the droplet size is much smaller than the levitation height h. Since the gas phase is moist air at moderate temperature and humidity, evaporation takes place under conditions when diffusion is important, in contrast to, e.g., the case of Ref. [13]. Because of the small system size, the unsteady effects and convective mass transfer are both weak, so the local vapor density distribution can be described by the steady diffusion equation. Diffusion around a droplet leads to fluid motion known as the Stefan flow. If density ρ of the air-vapor mixture is approximately uniform in space [18], the total partial density ρ_g of all components of air except water vapor decreases as the droplet surface is approached. Thus, there is diffusion of these components in the direction toward the droplet surface. However, since the components of air do not penetrate the liquid boundary, the diffusive mass transport of air has to be compensated by the macroscopically observable motion of the air-vapor mixture as a whole, i.e., the Stefan flow [18,19]. For an isolated evaporating spherical droplet the flow is spherically symmetric and the droplet acts as a source of strength Q, which by conservation of mass is

$$Q = -\frac{\rho_l}{\rho} 4\pi R^2 \frac{dR}{dt}, \tag{1}$$

where ρ_l is liquid density, t is time. The flow symmetry is broken in the presence of the wall. We propose that the reflection of the Stefan flow off the substrate is the mechanism of observed levitation phenomenon. We first illustrate this mechanism for a single droplet near a wall, see Fig. 2 (bottom right), using the Stokes flow approximation, appropriate since the Reynolds number is small ($\sim 10^{-3}$–10^{-2}). By the method of images, the solution is a combination of flows from the image source, Stokeslet doublet, and potential dipole [20], with induced velocity components

$$u_i = \frac{Q}{4\pi}\left[-\frac{r_i}{r^3} + \frac{6r_i r_2^2}{r^5} + 2h\left(\frac{\delta_{i2}}{r^3} - \frac{3r_i r_2}{r^5}\right)\right], \quad i = 1, 2, 3, \tag{2}$$

where r is the distance to the image and δ denotes the Kronecker delta. We note that since droplets are treated as point sources or sinks, deviations between theory and experiment are expected for larger droplets. Based on the above formula, the vertical component of the induced flow velocity at the location of the droplet ($r = 2h$) is $U = 3Q/(16\pi h^2)$. Assuming diffusion-limited evaporation [18], the droplet radius is expressed as $R^2 = R_0^2 - \alpha t$, where R_0 is the value at the start of the experimental recording for each data set. Substituting this classical law into Eq. (1) allows us to express Q (and thus U) in terms of the rate coefficient α found from the experimental recording of $R(t)$. The corresponding Stokes drag force, $6\pi\mu RU$, μ being the dynamic viscosity of moist air, should balance the force of gravity, leading to the following simple expression for the levitation height,

$$h = \frac{3}{4}\left(\frac{3\alpha\mu}{\rho g R}\right)^{1/2}. \tag{3}$$

This expression describes levitation height for an isolated droplet near the wall, so it is not directly applicable to our experiments in which an array of droplets is observed with spacing being of the same order of magnitude as the levitation height. For a given droplet in the array, interactions with the *images* of all other droplets contribute to the vertical force. To account for that interaction we introduce a correction to the Stokes force computed above for the isolated levitating droplet, leading to a correction factor, denoted by β, in the formula for h. This coefficient is in general a function of the array geometry, but for the purposes of our approximate model we take it to be constant. We estimate $\beta \approx 1.8$ using the nearest neighbor interaction approximation. The final result for levitation height can then be represented in nondimensional form as the following power law,

$$\hat{h} = \hat{R}^{-3/2}, \tag{4}$$

where $\hat{h} = h/R$, $\hat{R} = (2R/3)[\alpha\mu\beta^2/(2\rho g)]^{-1/3}$. The experimental data follow this power law as seen in Fig. 3 (top) for $\hat{h}$ above ~ 2 (note the condition $\hat{h} \gg 1$ used in the method of images). All quantities used in the plot, including β, are computed rather than obtained by fitting from the data. At $\hat{h} \sim 2$, a crossover to a different regime is observed. In this regime, the geometry is similar to the one used in classical studies of Leidenfrost droplets [12,13], so the dimensional distance between the droplet and the wall is the key parameter; this distance, scaled by R, is plotted in the inset of Fig. 3 (top) and follows the power law $\hat{h} - 1 \sim \hat{R}^{-2}$ (solid line, with the best agreement for smaller droplet-wall spacing). Evaporation into the narrow gap between the droplet and the wall has to be compensated for by axisymmetric gas flow in the gap. The pressure gradient associated with this flow is likely to be an important factor in droplet levitation, but developing mathematical models

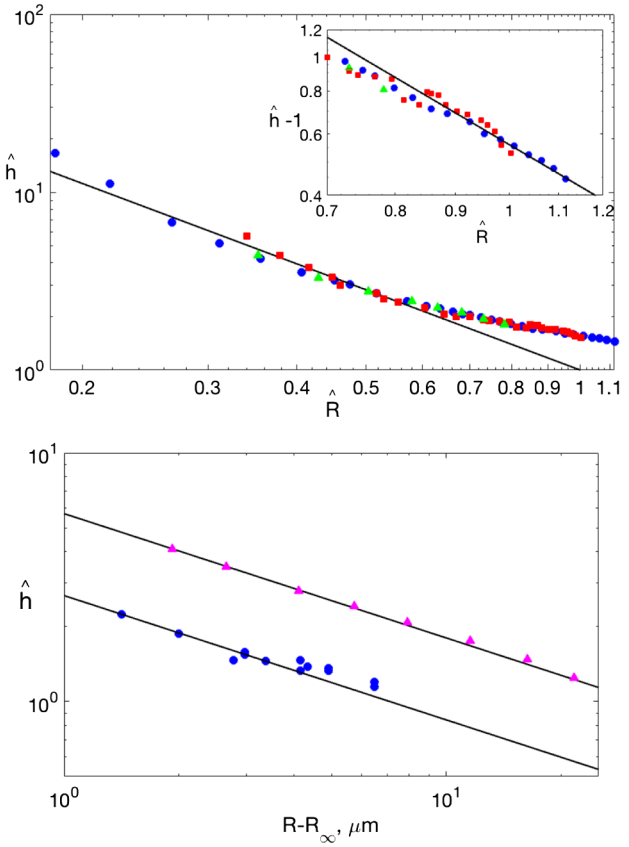

FIG. 3. (top) Comparison between experiments (three different droplets at $T_w = 85\,^{\circ}\mathrm{C}$) and theory [Eq. (4), solid line] for droplets levitating over a dry substrate; inset illustrates the power law corresponding to Leidenfrost-type situation at lower $\hat{h}$. The initial droplet radii are $R_0 = 8.6$ (circles), 7.8 (squares), and 6.0 μm (triangles). (bottom) Data for droplet levitation over liquid surface from our experiment (circles, $T_w = 84\,^{\circ}\mathrm{C}$, $R_\infty = 6.4$ μm) and the experiment of Kabov *et al.* [21] (triangles, $R_\infty = 22.1$ μm) both shown to follow the power law derived from Eq. (5).

for this configuration is beyond the scope of the present study.

While flow generated by the droplet images explains levitation, the Stefan flow around the actual physical droplets accounts for the repulsive interaction between them. It is this repulsion that provides a simple qualitative explanation for self-organization of droplets into regular structure, as seen in Fig. 2. There is no coalescence due to repulsion, but localization of all droplets within the dry patch is explained by the flow originating near the contact line; this flow has a component directed inward with respect to the dry part of the substrate.

Let us now use the insights from the new model and experiments to revisit the problem of levitation and self-organization of droplets over *liquid* surfaces. Fedorets *et al.* [3] obtained experimental evidence of upward flow of vapor-liquid mixture and suggested that the Stokes drag force generated by this flow is responsible for droplet levitation. They then conducted careful experimental measurements of mass loss of vapor and used these to estimate the velocity of the upward flow. However, their work fails to predict the experimentally observed preferential levitation height which exists under a wide range of conditions. We are now in position to resolve this contradiction.

In order to explain the dependence of levitation height on droplet size, consider how a condensing droplet affects otherwise uniform Stefan flow, of speed denoted by U_0, originating at the flat liquid-air interface. Since the Stefan flow velocity is proportional to the local gradient of the vapor density, $\nabla\rho_v$ [18], the key question is how this gradient is modified when a small condensing droplet is introduced. Such a droplet acts as a sink for the steady diffusion equation describing ρ_v solved with the boundary condition of constant vapor density, equal to the saturation value, at the flat interface. The solution of this problem is obtained by the method of images and leads to a modification of $\nabla\rho_v$ by $\sim Q/h^2$ due to the presence of the droplet. This leads to the condition of levitation of the form

$$6\pi\mu R\left(U_0 - \frac{\tilde{\beta}Q}{h^2}\right) = \frac{4}{3}\pi R^3\rho_l g, \tag{5}$$

with a fitting parameter $\tilde{\beta}$. Equation (5) suggests that if the scaled levitation height $\hat{h}$ is plotted versus $R - R_\infty$, R_∞ being the radius corresponding to very large values of $\hat{h}$, a new power law of the form $\hat{h} \sim (R - R_\infty)^{-1/2}$ should be observed for $R - R_\infty \ll R_\infty$. Figure 3 (bottom) clearly shows that this is indeed the case for both our new experimental data from a moving array of droplets (circles) and the previously published data for the localized cluster of levitating droplets (at a higher heat flux) from Kabov *et al.* [21]. These observations explain the unexpected similarity between the droplet behavior over dry surface and liquid-gas interface: in both of these the interaction of droplets with flat interfaces is of critical importance.

To summarize, we demonstrated experimentally for the first time that an ordered array of levitating droplets can form over a dry substrate rather than a liquid-gas interface. The relative height of droplet levitation in these experiments at h/R above ~ 2 follows the $R^{-3/2}$ power law, in very good quantitative agreement with a model we developed to describe droplet levitation in the Stefan flow created by their images. This leads us to propose Stefan flow as an important contribution to the mechanism of droplet levitation over liquid-gas interfaces as well. Comparison with experiments supports this conclusion.

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