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2019

Zhao, Y., Zhu, F., Zhang, H., New, T. H., Jin, L., & Yang, C. (2019). Triple condensate halo from a single water droplet impacting upon a cold surface. *Applied Physics Letters*, 114(18), 183703-. doi:10.1063/1.5080356

<https://hdl.handle.net/10356/105152>

<https://doi.org/10.1063/1.5080356>

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Cite as: Appl. Phys. Lett. **114**, 183703 (2019); doi: [10.1063/1.5080356](https://doi.org/10.1063/1.5080356)

Submitted: 7 November 2018 · Accepted: 20 April 2019 ·

Published Online: 6 May 2019

View Online

Export Citation

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ABSTRACT

Understanding the dynamics of water droplets impacting upon a solid surface is of importance from both fundamental and practical standpoints. While the impact dynamics of a water droplet upon a heated surface has been extensively studied, the impact characteristics of droplets upon a cold surface remain elusive. Here, we report the formation of a triple condensate halo observed during a water droplet impacting at low velocity upon a cold surface. Due to the interplay of droplet impact and vapor mass diffusion during the droplet spreading and cooling processes, two condensation stages occur, engendering this unique condensate halo with three distinctive bands. We further examine experimentally the effects of droplet impact velocity (in terms of Weber number) and substrate temperature on the three bands of condensate halo. We also provide scaling analyses to explain the experimental results. Our experimental investigation and theoretical analysis reported in this study reveal insights into the droplet impact dynamics and the associated condensation heat transfer.

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Liquid droplets impacting upon solid surfaces are ubiquitous in nature and have many industrial applications. Numerous experimental and theoretical works have been carried out, aiming at describing and predicting the kinetic behaviors of impacting droplets.^{1–5} Unraveling the physical mechanisms pertaining to the impinging, spreading, and recoiling/rebounding/splashing processes under various conditions is of significant importance in a variety of applications from ink-jet printing,^{6–8} spray cooling,^{9–12} and self-cleaning surfaces,^{13–16} to anti-icing in the aviation industry.^{17–21} However, the complex interplay of factors such as incident velocity, surface topography and temperature, thermophysical conditions of the ambient, and liquid properties makes droplets impacting on solid surfaces an intriguing and difficult topic, which remains inadequately understood till this day.

During the impact of a liquid droplet upon a smooth and chemically nonreactive substrate, mass, momentum, and energy are exchanged between the droplet and the surface. In particular, such exchanges can lead to unique droplet behavior, when the surface is either superheated ($T_b \gg T_{\text{boiling}}$) or subcooled ($T_b \ll T_{\text{melting}}$). When a droplet impacts upon a superheated surface with a temperature above a certain threshold, water gets evaporated so fast that the droplet essentially floats on vapor, and this phenomenon is referred to

as the Leidenfrost effect.^{22–24} On the contrary, when a droplet impacts upon a subcooled surface, heterogeneous ice nucleation occurs spontaneously at the water/solid interface.^{25–28} The contact line is pinned and the excess kinetic energy is therefore dissipated by constrained trampling instead of spreading.^{29–31} On the other hand, the scenario whereby the cold surface temperature falls between the freezing point and the dew point where only condensation occurs, has seldom been reported. Experimental characterization for exploration of its underlying mechanisms is thus needed.

In this work, we report our findings on the dynamic behaviors of a warm water droplet impacting upon a cold surface under the condition $T_{dp} > T_b > T_{ice}$, where T_{dp} , T_b , and T_{ice} denote the dew point, substrate temperature, and heterogeneous ice nucleation temperature, respectively. We discovered a unique triple halo pattern formed during the droplet impacting process. The formation of such a condensate pattern results from a two-stage condensation: the first occurs during the droplet spreading process, and then followed by the second during the droplet cooling process. Additionally, based on the interplay of droplet impact dynamics and vapor mass diffusion, we provide scaling analyses to elucidate the variations in the halo patterns with droplet impact velocity and surface temperature changes.

We carried out a visualization based experimental study to directly observe what happens when a water droplet impacts upon a cold substrate surface. Figure 1(a) shows a schematic of the main experimental components and setup used in this work. More information on the experimental details is provided in [supplementary material](#). The chamber is filled with N_2 gas at a room temperature of $T_\infty = 22 \pm 0.5^\circ\text{C}$. The droplet with a fixed volume of $10 \pm 0.2 \mu\text{l}$ (its typical radius is about $r_0 \approx 1.3 \text{ mm}$) and temperature T_∞ is produced and then released via gravity onto the substrate (with a temperature $T_b = 2.2 \pm 0.1^\circ\text{C}$). The corresponding Weber number is about $We \approx 13$.

FIG. 1. A triple condensate halo formed from a water droplet impacting upon a cold surface. (a) Schematic depiction of the experimental setup. (b) A condensate halo with three distinctive bands formed under the conditions of a $10 \pm 0.2 \mu\text{l}$ water droplet at $22 \pm 0.5^\circ\text{C}$ impacting (at a low velocity corresponding to a Weber number of $We \approx 13$) on a silicon wafer substrate with its temperature $T_b = 2.2 \pm 0.1^\circ\text{C}$.

Here, $We = 4\rho g h r_0 / \sigma$, ρ is the density of the fluid, g is the gravitational acceleration, h is the release height (from the droplet bottom to the substrate), and σ is the surface tension of water. Under such conditions, vapor from the evaporation of the impacting droplet gets condensed on the cold substrate and forms a halo pattern. The condensate halo is observed to consist of three distinctive bands as shown in Fig. 1(b). Though the incidence of condensation is expected when the warm droplet approaches the cold surface, the formation of such a halo with multiple bands is intriguing. In the literature, a single band condensate halo was observed due to the explosive release of water vapor upon recalescent freezing of a subcooled sessile droplet on a solid surface with its temperature below T_{ice} .³² Obviously, since the substrate temperature in our experiment falls in the range of $T_{dp} > T_b > T_{ice}$, the resultant phenomenon and its underlying mechanisms reported in this work are very different from those reported in Ref. 32.

Figure 2(a) shows a portion of the formed condensate halo (Multimedia view), with R_0 , R_1 , R_2 , and R_3 denoting the base radius of

FIG. 2. Part of the formed condensate halo with three bands under the conditions $We \approx 13$ and $T_b = 2.2 \pm 0.1^\circ\text{C}$. (a) An optical microscopy image with the variation of intensity along the radial direction. The inset shows the boundary of band 3 $|R_3 - R_2|$ by enhancing the local brightness and contrast. (b) Variation of the calculated condensate density along the radial direction, based on the optical image of (a), with R_0 , R_1 , R_2 , and R_3 denoting the locations where drastic changes occur. The inset shows a magnified plot of the selected rectangle indicated by the dotted line. Multimedia view: <https://doi.org/10.1063/1.5080356.1>

the droplet and the locations of three boundaries along the radial direction, respectively. Since the third band looks relatively darker, the inset shows its appearance by enhancing the local brightness and contrast. The widths of three bands are represented by band_1 $|R_1 - R_0|$, band_2 $|R_2 - R_1|$, and band_3 $|R_3 - R_2|$. The size of each band can be detected based on the sharp change in intensity as shown in [supplementary material Fig. S5](#).

The halo comprises micron-sized condensate droplets, and thus each band intrinsically represents the variation of condensed droplet size and number density. In particular, the boundaries indicate the locations where drastic changes in condensation occur. By calibrating a correlation between the image intensity and the condensate density (see [supplementary material Fig. S6](#)), we can extract information regarding the condensation variation along the radial direction as shown in [Fig. 2\(b\)](#). Therefore, the locations of R_1 , R_2 , and R_3 can be determined at locations where the condensate density drastically increases/decreases, depending on the nature of each band.

To provide more information on how a condensate halo is formed, we employed two synchronized cameras to observe the droplet impact behavior and the growth of the condensate halo from both side- and top-views. [Figure 3\(a\)](#) shows the dynamics of a water droplet impacting upon a cold surface from the side-view (Multimedia view), with $t = 0$ ms being the moment when the droplet touches the substrate. At $t \approx 10$ ms, the radius R reaches the droplet's maximal spreading^{33,34} with $R_{max} \sim r_0 We^{1/4}$, and the contact line starts to retract. At $t \approx 1$ s, the droplet reaches the equilibrium contact angle θ and its base radius R_0 . [Figure 3\(b\)](#) illustrates how the condensate halo grows as the droplet impacts upon the cold surface. Initially, the

pendent water droplet is surrounded by its own vapor, due to the diffusion-controlled evaporation. Of note is that when the droplet is brought close to the cold surface, the vapor pressure is unsaturated at chamber temperature T_∞ , but supersaturated at substrate temperature T_b . Once the droplet is deposited upon the cold surface, the excess vapor gets condensed on the cold surface. A condensate halo composed of three distinctive bands is formed due to the interplay of droplet spreading and vapor mass diffusion. Specifically, the formation of such a condensate halo consists of two sequential stages. The first-stage condensation occurs during the droplet spreading process, and the condensate extends from the droplet maximum spreading R_{max} to a certain distance outwards along the radial direction. The width of band 2 $|R_2 - R_1|$ is thus determined by the characteristic length of vapor mass diffusion during this stage, namely, $l_s \sim \sqrt{D t_s}$, with D being the diffusivity of water vapor and t_s the spreading time. It is worth mentioning that t_s is independent of the impact velocity³⁵ and scales with $\mu r / \sigma$, where μ is the dynamic viscosity of water. The second stage condensation happens during the droplet cooling process. Since the evaporation slows down when the droplet temperature is decreased, this stage lasts until the supply of vapor from the droplet evaporation stops, leading to $R = R_3$. The width of $|R_3 - R_0|$ is determined by the characteristic length of vapor mass diffusion within the second stage, $l_c \sim \sqrt{D t_c}$, with t_c being the characteristic time of droplet cooling. A direct energy balance analysis shows that t_c scales with $R_0^2 f_1(\theta) \frac{1}{\alpha}$, where $f_1(\theta) = \left(\frac{\sin \theta}{1 + \cos \theta}\right)^3 \left(1 - \frac{\sin \theta}{3 + 3 \cos \theta}\right)$ is a geometrical factor with θ being the equilibrium contact angle and α the thermal diffusivity of water. The derivation of the expression for t_c is provided in

FIG. 3. Droplet impact behavior and growth of the condensate halo. (a) Side-view images showing the dynamic behavior of an impacting droplet upon a cold surface under the conditions $We \approx 13$ and $T_b = 2.2 \pm 0.1$ °C. (b) Schematic illustrations showing growth of the condensate halo due to two-stage condensation. (c) Top-view images showing the growth of the condensate halo. Multimedia view: <https://doi.org/10.1063/1.5080356.2>

supplementary material. Figure 3(c) presents top-view images showing the growth of the condensate halo (Multimedia view). R_1 coincides with $R_{\max}$, as the condensate droplet falling within $R_{\max}$ is engulfed by the advancing contact line. The first two subfigures in Fig. 3(c) show that the second band of the condensate halo is formed between the droplet maximum spreading and the droplet recoiling, and band_2 $|R_2 - R_1|$ emerges right after the spreading process.

We carried out thorough experiments to characterize the triple condensate halo by examining the effects of the droplet impact velocity and the substrate temperature. Figure 4(a) shows the variation of each band width of the condensate halo with We . For the given droplet and substrate wetting angle conditions, the impact velocity has little

FIG. 4. (a) Variation in the size of condensate halo bands with Weber number, We (at fixed $T_b = 2.2 \pm 0.1^{\circ}\text{C}$). (b) Variation of the size of condensate halo bands with substrate temperature, T_b (at fixed $We \approx 13$).

influence on the local vapor concentration. Also, both the spreading time and the cooling time are not dependent on the impact velocity. Hence, two characteristic lengths of vapor mass diffusion, $l_s \sim |R_2 - R_1|$ and $l_c \sim |R_3 - R_0|$, do not show much dependence on We . Since $R_1 \approx R_{\max}$ scales with $rWe^{1/4}$, band_1 $|R_1 - R_0|$ and band_3 $|R_3 - R_2|$ increase and decrease with increasing We , respectively.

Figure 4(b) shows the variation of each band width of condensate halo with substrate temperature. It is known that the degree of supersaturation $S \equiv RH(T_{\infty})/RH(T_b)$ increases drastically with decreasing substrate temperature. Accordingly, as shown in supplementary material Fig. S7, the energy barrier of heterogeneous condensation nucleation, $\Delta G = \frac{16\pi M^2 \sigma^3}{3(RT \rho \ln S)^2} f_2(\theta)$,³² decreases with decreasing substrate temperature, where S is the degree of supersaturation, $f_2(\theta) = \frac{1}{2} - \frac{3}{4} \cos \theta + \frac{1}{4} \cos^3 \theta$ is the surface wetting factor, M is the water molecular weight, R is the idea gas constant, and ρ is the density of condensate. Therefore, band_2 $|R_2 - R_1|$ and band_3 $|R_3 - R_2|$ increase with decreasing substrate temperature T_b , while the variation of band_1 $|R_1 - R_0|$ with T_b is insignificant.

In summary, we reported an interesting phenomenon that a triple condensate halo is formed when a warm water droplet impacts upon a cold surface with the surface temperature between the dew point and the freezing point of water. Based on our experimental results and scaling analyses, we have shown that this condensate halo is formed due to interplay of the droplet impacting dynamics and the vapor mass diffusion. In particular, two condensation stages lead to the formation of the condensate halo that has a unique three-band feature with distinctive boundaries. Our systematic experimental studies reveal the characteristics of the triple condensate halo, specifically showing the dependence of the sizes of the three condensate halo bands on the droplet impact velocity and the substrate temperature. Such dependence can be explained by the scaling analyses presented in this work. From a broader perspective, this work presents an important advancement in our understanding of low velocity water droplet impacts with condensation phase changes. We envisage that such an intriguing condensation behavior could find a range of applications in condensation heat transfer related processes.

See supplementary material for experimental details, supplementary figures 1–7, and a derivation of the droplet cooling time.

We acknowledge the financial support from the Ministry of Education of Singapore via Tier 2 Academic Research Fund (MOE2016-T2-1-114) and the Nanyang Technological University Ph.D. Scholarship to F.Z. and H.Z.

The authors declare no competing financial interest.

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