

Electrowetting-Controlled Dropwise Condensation with Patterned Electrodes: Physical Principles, Modeling, and Application Perspectives

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Patterning the wettability of solid surfaces is a successful strategy to control the dropwise condensation of vapor onto partially wetting solid surfaces. The condensation of water vapor onto electrowetting-functionalized surfaces is studied with structured co-planar electrodes. A detailed analysis of the experimental distribution of millions of drops reveals that despite the presence of contact angle hysteresis and the occurrence of random drop coalescence events, the preferential drop positions closely follow the numerically calculated local minima of the electrostatic energy for variable drop size. Even subtle transitions between competing preferred locations are properly reproduced by the model. Based on this quantitative understanding of the condensation patterns, a series of important follow-up steps that need to be taken to demonstrate a reliable performance gain in various applications focusing in particular on enhanced heat transfer is discussed.

1. Introduction

The condensation of water vapor onto solid surfaces is integral to many natural processes including dew formation^[1] and fog harvesting by animals, like the Namib Desert Beetle,^[2,3] *Litoria caerulea*, a green tree frog native to Australia and New Guinea,^[4] and Cribellate spider,^[5] and plants, such as the Namib desert plant *Trianthema hereroensis*, and cacti like *Opuntia microdasys*.^[5] Water condensation is also intrinsic to various technological applications like fog harvesting,^[6–8] seawater desalination,^[9] and heat exchangers for power generation^[10] and

refrigeration.^[11,12] In all cases, efficient condensation and removal (or “collection”) of the condensed liquid is essential. The entire cycle of condensation consists of a series of steps, namely the nucleation of liquid on an initially dry solid surface, the subsequent growth of the liquid phase in form of droplets (or a film), and finally the removal of the condensate such that a subsequent generation of drops can nucleate. At first glance, hydrophilic surfaces may seem the most natural choice to promote condensation. Yet, it has been long known that plain hydrophilic surfaces are actually not the best choice because they promote the formation of thermally insulating condensed liquid films, which compromises condensation (for a review: see ref. [13]).

Hence, it is advantageous to use partially wetting solid surfaces where condensing vapor forms discrete drops that leave parts of the solid surface in direct contact with the to-be-condensed vapor. As these discrete drops are removed, they expose even more barer surface for subsequent generation of condensing drops. Like in the case of biological or technological fog harvesting surfaces, efficient removal of the condensate drops is therefore essential for the overall performance of the system.

Throughout recent years, various efforts have been made to optimize dropwise condensation and the subsequent removal of drops by efficient manipulation and transport using suitable topographical and chemical surface patterns.^[14–23] Such patterns generate an energy landscape in which condensing drops initially form either at random locations or at preferred hydrophilic nucleation sites. As drops grow, they experience the imprinted gradients in wettability, hit geometric boundaries, and coalesce with other drops. In either case, the original configuration of the drop becomes unstable and the drop moves toward a location of lower energy. Examples of such surface modifications include surfaces with alternating hydrophobic and hydrophilic stripes,^[14–16] superwetting Janus membranes,^[8] surfaces with conical geometries,^[3] superhydrophobic surfaces with grooves^[17] or nanostructures,^[18–20] and liquid-infused surfaces.^[21–23] The resulting drop displacements are either driven entirely by capillary forces or are assisted by gravity in case of vertically oriented condenser surfaces. In all cases, drops only move once the driving forces overcome the inherent pinning due to microscopic heterogeneities,^[24] as quantified by the contact angle hysteresis $\Delta\cos\theta = \cos\theta_r - \cos\theta_a$. Here, θ_r and θ_a are the receding and advancing contact angles. Accordingly, the need for efficient removal of the condensate drops has generated

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considerable interest in surfaces with low contact angle hysteresis such as superhydrophobic and liquid-infused surfaces for heat transfer applications with dropwise condensation.

The approaches described above all rely on passive wettability patterns imprinted onto the solid surface upon fabrication. In contrast, electrowetting (EW) allows for active tuning of the wettability and controlled transport of drops of conductive liquids, such as water, on partially wetting hydrophobic surfaces.^[25–28] The capacitive coupling between the drop(s) and suitably structured co-planar electrodes on the substrate that are covered by a thin hydrophobic polymer layer allow efficient control of the local wettability above the activated electrodes.^[28,29] Drops that are large compared to the width of a gap between two coplanar electrodes, preferentially align on the center of the gap where the minimum of the electrostatic energy for the droplet-dielectric-electrode system is located. As usual in EW, this minimum of the electrostatic energy $E_{el} = -C_{tot}U^2/2$ corresponds to the maximum of the total capacitance between the drop and the electrodes.^[28] Such electrically controlled alignment of drops was exploited to demonstrate controlled capture, release, and steering of rolling drops on inclined planes,^[30] as well as manipulation of drops in microfluidic two phase flow systems.^[31] The idea of manipulating condensing drops by EW was first explored by Kim and Kaviani,^[32] Baratian et al.^[33] later combined the basic EW ideas to study for the first time directly the condensation of water vapor onto EW-functionalized surfaces. For the specific case of parallel interdigitated electrodes aligned along the direction of gravity, they showed that the condensate drops migrate to and align along the drop size-dependent minima of the (1D) electrostatic energy landscape perpendicular to the electrodes. Eventually this results in a periodic pattern of drops. Furthermore, the increased coalescence events during EW-induced migration lead to faster growth of drops. Up to this point, the condensation on EW-functionalized surfaces is very similar to condensation on chemically patterned substrates. Beyond that, however, EW with AC voltage also results in reduced contact angle hysteresis^[34] culminating in shedding of on average smaller drops, as compared to the reference case without EW.^[33] The faster growth of condensate drops and the smaller critical shedding radius result in higher shedding frequency which can be beneficial for heat transfer applications.^[13,33] A series of follow-up studies confirmed these basic original observations regarding the evolution of the drop distribution for straight interdigitated electrodes.^[35–39]

Experiments with more complex electrode geometries with zigzag-shaped edges resulted in preferential alignment of the drops not only perpendicular but also along the direction of the electrodes, in qualitative agreement with expectations.^[40] That study also indirectly inferred an increased heat transfer from the volume of shedded drops under AC-EW as extracted from video microscopy images. Overall, the acquired understanding suggests that it should be possible to optimize the performance of EW-controlled condensation in heat transfer and other applications by systematically varying electrode geometries and/or electrical excitation patterns. However, a purely empirical optimization of electrode shapes is very time consuming and costly. Therefore, it is essential to extend the existing electrostatic models for EW to arbitrary electrode geometries, and to demonstrate their efficacy in capturing the complex evolution of drop distribution patterns to enable electrode optimization in silico prior to experimental testing in the future.

In the spirit of establishing electrostatic models for EW as a platform for optimizing EW controlled dropwise condensation, the core of the present work consists of a detailed comparison of the experimentally observed evolution of the distribution of several tens of millions of drops with sizes between 4.3 and 2000 μm with the predictions of a numerical model based on the drop size-dependent minimization of the electrostatic energy. Experiments and numerical calculations are carried out for the specific case of interdigitated electrodes with zigzag shaped edges of variable length. Compared to the originally studied interdigitated electrodes with straight edges,^[33] this geometry offers the additional advantages that drops are transported not only perpendicular to the electrodes but also along them in the downward direction along with gravity. This sweeps a larger fraction of the surface clean and increases the size of the drops—which facilitates gravitationally driven shedding. This potential is expected to be particularly effective if combined with smart electrical actuation schemes as well as additional electrodes that will ultimately enable a “conveyor belt”-like active transport of drops. The numerical calculations reproduce the experimental observations, including a series of subsequent transitions of preferred drop positions as a function of size. The previously proposed simple analytical model by ‘t Mannetje et al.^[41] also reproduces the qualitative behavior but underestimates electrostatic energies and forces for small drops. Following the discussion of these results, we evaluate the present status of the field and discuss aspects that we consider essential for the development of EW-controlled condensation from a physical phenomenon toward a technologically relevant application.

2. Experimental Section

2.1. Experimental Aspects

The present condensation experiments were performed in the same homemade experimental setup (**Figure 1a**) that was used in our previous studies.^[33,40] The setup consists of a condensation chamber with two inlets at the bottom and an outlet through a fine grid of holes for vapor at the top side. The transparent sample was mounted vertically on one of the side walls and cooled from the back by cooling water (11.5 °C) from a commercial cooler (Haake-F3-K, Thermo Fisher Scientific). The sample was back-illuminated with an LED pad (MB-BL305-RGB-24-Z, Metabright) and imaged from the opposite side through an indium-tin-oxide (ITO)-coated heated window with a camera (Point Grey, FL3-U3) through a 20× zoom lens (Z125D-CH12, EHD). The resulting field-of-view is $\approx 10 \times 7.5$ mm (see Movie S1, Supporting Information). The temperature inside the chamber was measured by several thermistors (TCS651m AmsTECHNOLOGIES and Thorlabs TSP-TH) using a National Instruments data acquisition (DAQ) card and Labview and with the Thorlabs TSP01 Application. Thermistors were located at the vapor inlet, in the vapor close to the sample surface, at the vapor outlet, in the coolant behind the sample, in the heated water on the hot plate, and in the ambient air.

Deionized water (Millipore Synergy UV, 18.2 M Ω cm) was heated on a hot plate (RCT Basic, IKA labortechnik). Ambient air was blown through the water using an aquarium pump (0886-air-550R-plus, Sera) at a flow rate of 3.5 l min⁻¹, as monitored by a

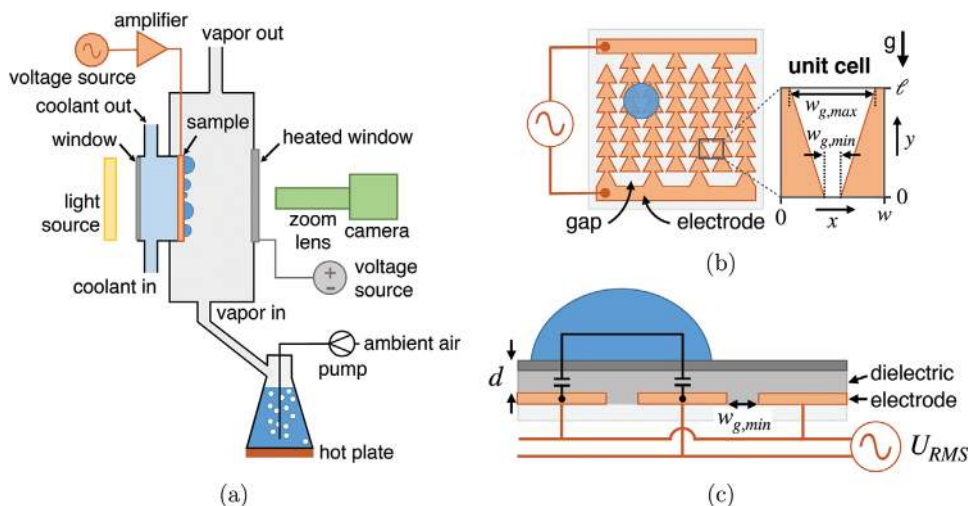


Figure 1. Experimental setup (not to scale). a) Schematic of vapor generator, condensation chamber, cooled sample stage, and optical setup. b) Top view of vertically oriented sample with zoomed view of unit cell of electrode pattern. c) Cross-sectional view of a condensed drop on the substrate.

flow meter (AWM5101VN flowmeter, Honeywell). The condensation chamber was initially kept dry with a steady flow of dry Nitrogen. At the start of an experiment, the humidified air at a relative humidity $>99\%$ and flow rate of 3.5 l min^{-1} was guided into the condensation chamber at the bottom of the chamber at a temperature of $42 \text{ }^\circ\text{C}$. The subcooling of the surface was kept constant at $\approx 30.5 \text{ }^\circ\text{C}$ throughout all experiments.

The recorded images were analyzed using a home-built image analysis routine in MATLAB to evaluate the center locations and radii of all the condensing drops (Figure S2, Supporting Information). The smallest drop size R detectable using this method was $R_{\min} \approx 4.3 \text{ }\mu\text{m}$.

The interdigitated zigzag electrodes were fabricated using photo-lithography on a glass substrate. The electrodes were subsequently coated with a $2 \text{ }\mu\text{m}$ thick dielectric layer of Parylene C (PDS2010, SCS Labcoater) using chemical vapor deposition (CVD), and an ultra thin top hydrophobic polymer coating (CytopTM, Asahi Glass Co., Ltd.) using a dip-coating procedure. For the experiments and simulations reported herein, interdigitated electrodes with zigzag-shaped edges were used (Figure 1b,c). As in ref. [40], the minimum and maximum width of the gap between adjacent electrodes were kept fixed at $w_{g,\min} = 50 \text{ }\mu\text{m}$ and $w_{g,\max} = 250 \text{ }\mu\text{m}$, and three different lengths ℓ of 500, 1000, and 3000 μm were tested. For AC-EW, an amplified electrical signal of rms amplitude between $U_{\text{RMS}} = 100 - 150 \text{ V}$ and a fixed frequency of $f = 1 \text{ kHz}$ was used using a function generator (Agilent 33220A) and voltage amplifier (Trek PZD700A). Advancing and receding contact angles at zero voltage were $\theta_a = 114^\circ$ and $\theta_r = 105^\circ$, implying a contact angle hysteresis of $\Delta\theta_0 = 9^\circ$. Upon applying an EW voltage of $U_{\text{RMS}} = 150 \text{ V}$, these values decrease to $\theta_a(150\text{V}) = 95^\circ$ and $\theta_r(150\text{V}) = 91^\circ$ following Lippmann's equation. This corresponds to a decreased contact angle hysteresis of $\Delta\theta(150\text{V}) = 4^\circ$, as expected for EW with AC voltage.^[34] This corresponds to a reduction of the pinning force by more than a factor 2 ($\Delta\cos\theta_0 = 0.15 \pm 0.02$ and $\Delta\cos\theta(150\text{V}) = 0.06 \pm 0.01$).

2.2. Numerical Aspects

To explain the experimental observations, a 3D numerical model that allows to calculate the electrostatic energy of a drop was developed as a function of its size and the (x, y) position of its center of mass within the unit cell of the electrode pattern (see zoomed view in Figure 1b). Since $\theta(150\text{V}) \approx 90^\circ$, we represent the drop by a simple hemisphere with radius R and with a fixed electrical conductivity (10^{-5} S m^{-1}) that guarantees (for all practical purposes) complete screening of the electric field from the inside of the drop. Note that this hemispherical approximation neglects slight EW-induced distortions of the drop shape (see below). Yet, earlier simulations showed that this merely leads to a minor underestimation of the electrostatic trapping strength for rather weakly deformed drops as in the present experiments.^[42]

The calculation of $E_{\text{el}}(x, y; R)$ starts with the calculation of the distribution of the electrostatic potential $\phi(x, y, z)$ within a 3D computational domain consisting of the electrodes, the dielectric layer, a single water drop of radius R , and the surrounding air. ϕ and the free charge density ρ_e is related according to the Poisson equation as

$$\nabla^2\phi = -\frac{\rho_e}{\epsilon_0\epsilon} \quad (1)$$

Here ϵ_0 is the permittivity of free space, and ϵ is the relative permittivity of the computational domain. ρ_e can be related to the current density \vec{j} using the charge conservation equation as

$$\frac{\partial\rho_e}{\partial t} = -\nabla\cdot\vec{j} = \nabla\cdot\sigma\nabla\phi \quad (2)$$

where σ is the electrical conductivity of the computational domain. Taking the time derivative of Equation (1), and subsequently substituting Equation (2) in it, we get a second order partial differential equation in ϕ :

$$\nabla^2 \phi = -\nabla \cdot \left(\frac{\sigma}{\epsilon_0 \epsilon} \nabla \phi \right) \quad (3)$$

Considering a sinusoidal electrical potential $\phi = \phi_0 \Re[e^{i\omega t}]$, and subsequently, considering its time derivative $\dot{\phi} = \phi_0 \Re[i\omega e^{i\omega t}]$, Equation (3) can be rewritten as

$$\nabla \cdot \left[\left(\epsilon_0 \epsilon - i \frac{\sigma}{\omega} \right) \nabla \phi \right] = 0 \quad (4)$$

Equation (4) was solved numerically in COMSOL Multiphysics (version 5.4) using the finite element method for a fixed voltage (amplitude) of 150 V and frequency of 1 kHz. The discretization or element order of modeling domains was varied between quadratic and the fifth-order in order to achieve the desired accuracy. Since the drop size in our experiments varies from a fraction of the width of a unit cell at early stages to drops covering several adjacent electrodes during later stages, the computational domain was chosen to be sufficiently large to cover the entire drop as well as the immediately adjacent electrodes. (In practice, several domain sizes were chosen for different ranges of drop sizes in order to reduce computational efforts.) The geometries of electrodes and dielectric films were chosen according to the experiments. Dirichlet boundary conditions (fixed electrostatic potential) were imposed on the electrode surfaces; Von Neumann conditions (zero electric field in normal direction) were applied on all other boundaries. Figure S3, Supporting Information shows a typical view of a computational domain along with the resulting potential distribution for a specific drop configuration. As mentioned above, these calculations were repeated for 200 values of the drop size R between 0 and 900 μm , and for each drop size at 30×30 (large R) or 30×60 (small R) spaced positions within the unit cell. The vertical resolution was thus $300 \mu\text{m}/30 = 10 \mu\text{m}$, the horizontal resolution varies from $500 \mu\text{m}/60 \approx 8 \mu\text{m}$ to $3000 \mu\text{m}/30 = 100 \mu\text{m}$. (For symmetry reasons, it was sufficient to vary

the drop positions only within half of a unit cell; see grey shaded area in Figure S3, Supporting Information).

After numerical evaluation of $\phi(x, y, z)$ for all allowed drop sizes and (x, y) -location within the unit cell, the total electrostatic energy of the entire system was calculated as

$$E_{\text{el}}(x, y; R) = - \int_v \frac{1}{2} \vec{E} \cdot \epsilon_0 \epsilon \vec{E} dv = - \frac{1}{2} \int_v \epsilon_0 \epsilon \left(\left| \frac{\partial \phi}{\partial x} \right|^2 + \left| \frac{\partial \phi}{\partial y} \right|^2 + \left| \frac{\partial \phi}{\partial z} \right|^2 \right) dv \quad (5)$$

where $\vec{E} = -\nabla \phi$ is the electric field, and the integration represents the volume integral over the entire computational domain.

In the representation of the electrostatic energy landscapes later on (Figure 5), symmetries and periodicities were used to extend the energy landscapes beyond a single unit cell for a more intuitive representation. Finally, note that Equation (4) contains both dielectric and purely conductive contributions. However, for the conductivity of pure water and for the applied (low) frequency, the ionic current dominates the displacement current toward screening the electric field (also see ref. [33]).

3. Results

3.1. Evolution of Breath Figures

As apparent at first glance, the condensate drops form a pattern (breath figure) with well-defined periodicities along both the lateral (x -) and vertical (y -) directions upon condensation onto surfaces with zigzag interdigitated electrodes (Figure 2a–c). This is in sharp contrast to breath figures with straight interdigitated electrodes under AC-EW, where no periodicity along the y -direction was found.^[33] While these observations have qualitatively been reported before,^[40] a closer look at the representative Figure 2d–i reveals a number of additional details: Initially, the small condensate drops are essentially randomly

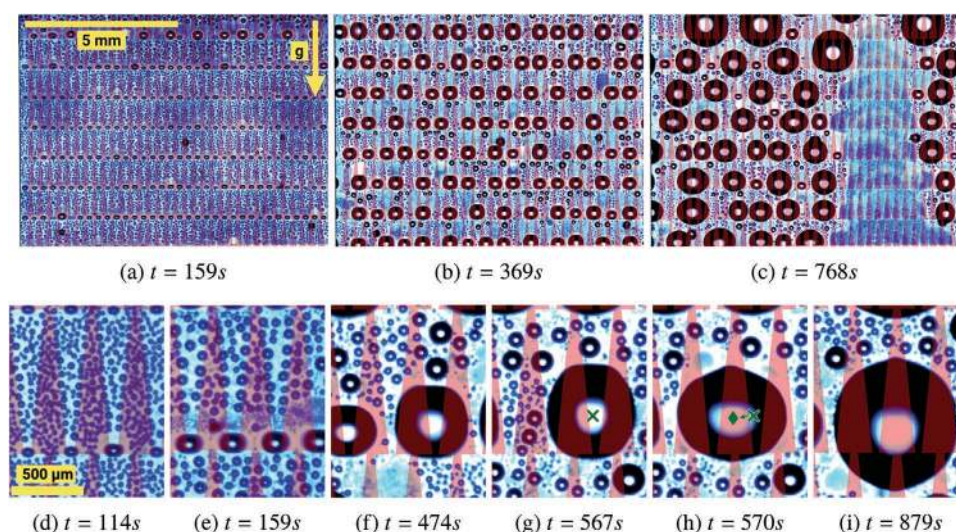


Figure 2. Top view of condensed droplets on a vertically mounted substrate ($l = 1000 \mu\text{m}$). a–c) Full field of view for $t = 159, 369, 768 \text{ s}$ illustrating alignment and growth of condensing drops. d–i) Zoomed view of ≈ 3 unit cells for times as indicated. Note the vertical and horizontal shift of the center of the drops with increasing size. (Transparent ITO electrodes are superimposed in red.)

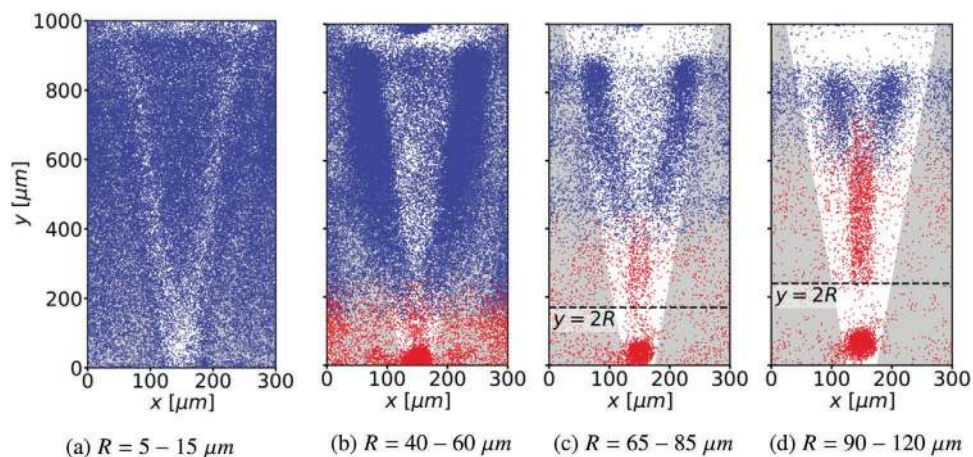


Figure 3. Center locations of all drops projected into single unit cell and binned to size ranges as indicated ($l = 1000 \mu\text{m}$). Drops with $R > 0.5w_g(y)$ are shown in red. $w_g(y)$ is the y -dependent gap width ranging from $w_g(0) = w_{g,\text{min}}$ to $w_g(l) = w_{g,\text{max}}$.

distributed; however, as the drops grow and begin to coalesce, they align parallel to the electrode edges, with a slight preferential displacement toward the gap centers (Figure 2d,e). Simultaneously, the drops closer to a gap minimum (i.e., at $(x = w/2; y = 0)$ in Figure 1b) are pulled down toward that minimum and typically grow on their way by coalescence with other drops (Figure 2d,e). As we will see below, drops at these ‘gap minima’ are trapped in electrostatic energy minima; as these continue to grow, their lower edge remains close to $y = 0$, whereas their center gradually moves upward (Figure 2d–g). These trapped, growing condensate drops dominate the visual appearance of the breath figures on a macroscopic scale (Figure 2a,b). Interestingly, upon reaching some critical size, the center of mass of these trapped drops suddenly translates horizontally from being centered on the gap between two adjacent electrodes to being centered on an electrode (see transition cross marker to diamond marker in Figure 2g,h). Upon growing further, the center of the trapped drops shifts slightly downward (Figure 2i). Eventually, at much larger radii, drops shed at a critical radius R_{shed} under the influence of gravity when the individual drop weight exceeds the electrical trapping force and contact angle hysteresis (Figure 2c).

For a statistical analysis of the distribution of condensate drops, we project the drop centroid locations of all drops within the field of view onto a single unit cell of the electrode pattern (Figure 1b) using a mapping procedure that takes into account the optical distortion of the imaging system (see Figure S2, Supporting Information). **Figure 3** shows the resulting spatial distribution of the drops within the unit cell for $l = 1000 \mu\text{m}$ binned into ranges of $R = 5\text{--}15$, $40\text{--}60$, $65\text{--}85$, and $90\text{--}120 \mu\text{m}$, where each data point represents the location of a drop center at a particular moment in time.

While the distribution of the smallest drop sizes ($5\text{--}15 \mu\text{m}$; Figure 3a) is almost random, somewhat larger drops ($40\text{--}60 \mu\text{m}$) preferentially align along the inclined edges of the electrodes (Figure 3b). As the drops coalesce and grow further, they gradually move from the electrode edges toward the gap center (Figure 3b–d). Drops with a diameter that exceeds the local width $w_g(y)$ of the gap, that is, drop with a critical size $R > 0.5w_g(y)$ (red data points) are preferentially found in the center of the gap

rather than along the electrode edges (Figure 3c,d), giving rise to a peculiar bi-modal distribution of the drops (Figure 3c). This bi-modal spatial distribution of drops is unique to the converging electrode geometry. In contrast, for straight electrode edges with a constant gap width, the drop distribution is always uni-modal (i.e., the drops of equal size align either on both sides of the gap center or along the gap center).^[33] The larger the drop size under consideration, the larger the fraction of drops with $R > 0.5w_g(y)$. Hence, the largest drops are again largely centered on the gap (red dots in Figure 3d), concomitant with a depletion of drops from the electrodes including their edges. The evolution of the spatial distribution of the condensate drops (Figure 3b–d) with increasing drop size is thus reminiscent of a “zipper-like” effect. The cluster of data points in the vicinity of the gap minimum always represent electrically trapped droplets (Figure 3b–d). Furthermore, Figure 3c,d clearly show that the strong electrical force sweeps the drops within a distance of characteristic length scale $\approx 2R$ above the gap minimum (note the relative lack of droplets over this region) creating the bigger trapped droplet which continues to grow upward.

Another interesting series of transitions is revealed by plotting the correlation between average drop size R and the lateral position of their center of mass (**Figure 4a**). For $R > 0.5w_{g,\text{max}}$, most drops are preferentially aligned along the gap center ($x = 150 \mu\text{m}$). However, this gap-centered alignment of the condensate drops does not persist as the drops grow further. At another critical size R_1 ($\approx 320 \mu\text{m}$ for the present $l = 1000 \mu\text{m}$ electrode), drops on average undergo a transition from being centered on the middle of the gaps to being centered on the middle of the adjacent electrodes, as already described for the specific individual drop in Figure 2g,h. Beyond that, a series of additional transitions back and forth the centers of gaps and electrodes are seen at critical radii R_2, R_3, R_4 , yet increasingly faint due to decreasing numbers of larger drops. The positions of the dashed horizontal lines emerge from the numerical model (see below). The same series of transitions are also observed for the other electrode geometries with $l = 500 \mu\text{m}$ and $l = 3000 \mu\text{m}$ (see Figure S4, Supporting Information).

In order to further visualize the spatial evolution of the trapped drops, Figure 4b shows the projected vertical (y -

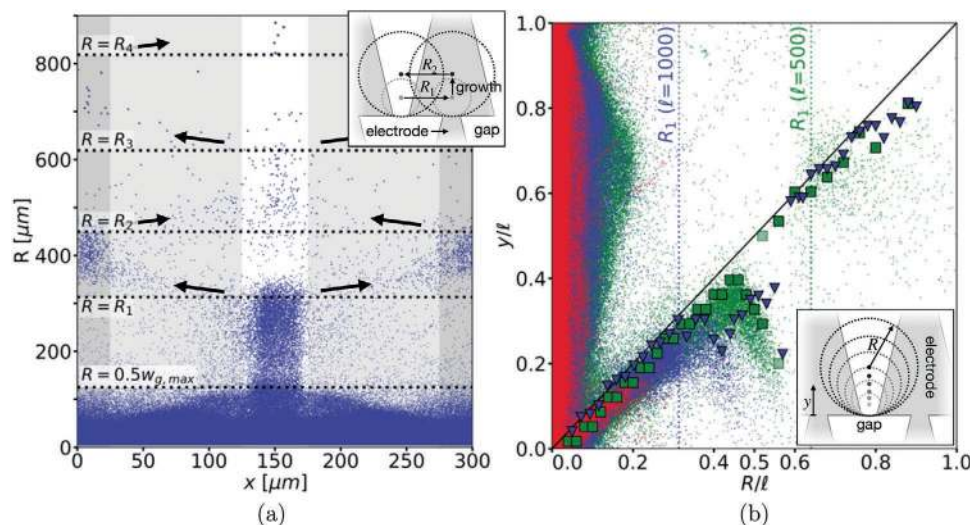


Figure 4. a) Horizontal and b) vertical distribution of experimental drop centers (small dots) within a unit cell for variable drop radius R . a) Horizontal dotted lines indicate critical radii for first central alignment ($0.5w_{g,max}$) and subsequent transitions ($R_1...R_4$) from preferred alignment on gap center ($x = 150 \mu\text{m}$) and electrode center ($x = 0$ or $300 \mu\text{m}$), as extracted from Figure 6 (data for $l = 1000 \mu\text{m}$). Bright and light shaded regions indicate the minimum and maximum gap widths $w_{g,min}$ and $w_{g,max}$. Inset: illustration of horizontal transitions of drop center upon growth. b) Normalized vertical position of drop center for all electrode sizes (green: $l = 500 \mu\text{m}$; blue: $l = 1000 \mu\text{m}$; red: $l = 3000 \mu\text{m}$) versus normalized drop size. Large symbols: vertical position of electrostatic energy minimum versus drop size extracted from numerical calculations (see Figure 5): green squares: $l = 500 \mu\text{m}$; blue triangles: $l = 1000 \mu\text{m}$. Solid line: geometric approximation $y = R$. Inset: illustration of geometric shift of drop center for bottom of drop pinned at minimum gap.

locations of all drops normalized by the electrode length (y/l) versus R/l for all three electrode designs ($l = 500, 1000, 3000 \mu\text{m}$). The tail developing from the gap minimum ($y/l = 0$) represents the vertical locations of the trapped drops. Initially, the vertical locations of these trapped drops satisfy $y \approx R$ (solid black line in Figure 4b), as previously observed from Figures 2 and 3. Although for the $l = 3000 \mu\text{m}$ electrode (red data points) the trapped drops stay aligned at $y \approx R$ till gravity-driven shedding ($R_{shed}/l \approx 0.3$), for the $l = 1000 \mu\text{m}$ (blue) and $l = 500 \mu\text{m}$ (green) electrodes the drops subsequently deviate from the line $y = R$ as these continue to grow by coalescence (Figure 4b). For the $l = 1000 \mu\text{m}$ electrode, this deviation begins immediately after the first lateral transition ($R_1/l \approx 0.32$), while for the $l = 500 \mu\text{m}$ electrode this occurs well before the first lateral transition at $R/l \approx 0.5$ (Figure 4b). Interestingly, once the trapped drops grow bigger, they realign again following $y \approx R$ for both the $l = 1000 \mu\text{m}$ and $l = 500 \mu\text{m}$ electrodes above $R/l \approx 0.6$ (Figure 4b). Note that for both $l = 1000 \mu\text{m}$ and $l = 500 \mu\text{m}$ electrodes, the deviation of the trapped drops from the line $y = R$ occurs well below the critical shedding radius ($R_{shed} \approx 1 \text{ mm}$); hence, gravity is not the cause of the deviation.

3.2. Electrostatic Energy Landscape Controls the Evolution of Breath Figures

The details of the drop distributions described above can be understood by considering the 2D electrostatic energy landscape ($E_{el}(x, y)$) emerging from our numerical calculations. Figure 5 illustrates the evolution of these energy landscapes for electrodes with $l = 1000 \mu\text{m}$ (Figure 5a–e) and $l = 500 \mu\text{m}$ (Figure 5f–j) for a series of drop sizes as indicated in the figure. As noted above, the energy landscape is by construction mirror

symmetric along the center of the gap. For the smallest drops, the energy landscape is rather flat with shallow valleys along the electrode edges that become deeper upon approaching the gap minimum at $y = 0$ (Figure 5a). For larger drops, (Figure 5b,c), the two separate minima along the electrode edges first merge into one minimum centered at $x = w/2, y \approx R$, leading to a coexistence of a single minimum close to the gap center in the lower parts of the unit cell and two valleys in the upper parts. Such variation in the $E_{el}(x, y)$ landscape is consistent with the drop distribution shown in Figure 3b–d and its “zipper-like” evolution with increasing drop size. The y -coordinate of the central minimum gradually shifts toward larger y for sizes comparable to the trapped drops, consistent with the solid $y = R$ line in Figure 4b.

As the drop size approaches $300 \mu\text{m}$ for $l = 1000 \mu\text{m}$, the electrostatic energy minimum eventually moves from the gap center to the electrode center (Figure 5d,e). We can predict the drop radius for this and subsequent lateral transitions by calculating the E_{el} for a drop located either at the gap center or at electrode center, for a range of drop radii (Figure 6). For small drop sizes ($R < 320 \mu\text{m}$), the total electrostatic energy is smaller when the drop is located at the gap center (black solid line in Figure 6) than when located at the electrode center (red solid line). As the drop size increases further, the location of the lowest electrostatic energy moves alternately between the electrode and the gap centers (compare the relative variations between the black and red solid lines in Figure 6). The characteristic radii at which these transitions occur, R_1, R_2, \dots, R_n in Figure 6 are shown as horizontal dashed lines in Figure 4a, and provide a good description of the transitions observed experimentally.

Tracing the position of the global energy minimum along the y -direction reveals that the drop center indeed moves upward

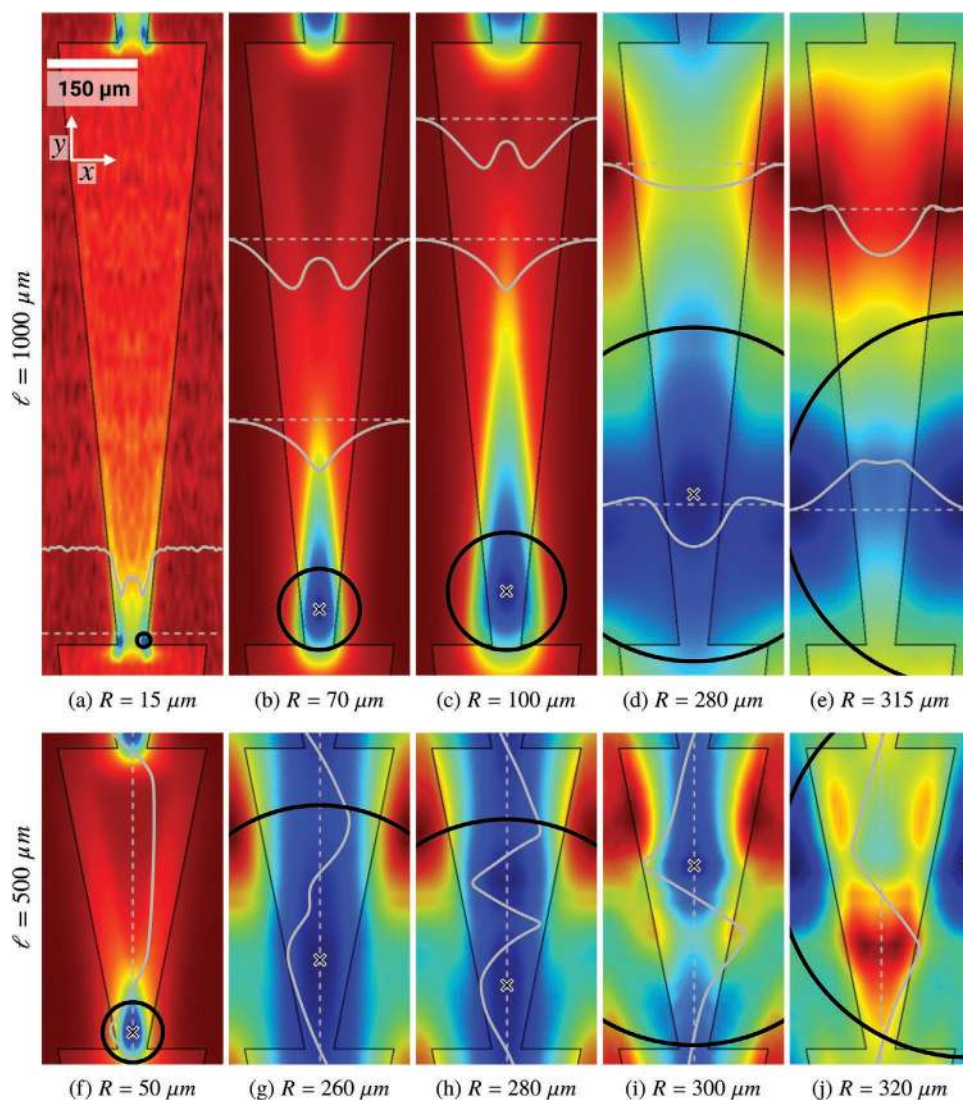


Figure 5. False color representation of 2D energy landscape (blue=low; red=high energy) versus (x, y) position of drop center for various drop sizes as indicated. Top row a–e): $l = 1000 \mu\text{m}$; bottom row f–j): $l = 500 \mu\text{m}$. Grey lines: cross sections through electrostatic energy landscapes along dashed lines; crosses and black circles: centers and edges of drops at minimum energy configuration. (Color scales and cross sections are individually rescaled for optimum visual contrast; Data in (a) are somewhat compromised by numerical noise.)

with increasing drop size following slightly below the line $y = R$, as shown in Figure 4b. The numerical results (squares and triangles in Figure 4b) reproduce the pertinent experimental observations. In some cases correlations with lateral transitions of the drop position can be observed.

For the shorter unit cell ($l = 500 \mu\text{m}$), the evolution of the energy landscape is qualitatively similar. Nevertheless, the two situations cannot be mapped directly onto each other. For instance, unlike the long electrodes, we find for $l = 500 \mu\text{m}$ that the energy minimum in the gap center splits up into two distinct local minima as the drop diameter becomes comparable to l between $R \approx 250\text{--}300 \mu\text{m}$ (Figure 5g–i). This leads to a distinct transition of the drop position along the y -direction for $R = 280 \rightarrow 300 \mu\text{m}$ (Figure 5h,i), while the drop remains laterally centered on the gap. This transition is indeed observed in the experiments with short ($l = 500 \mu\text{m}$) electrodes (Figure S4,

Supporting Information) but not for $l = 1000 \mu\text{m}$, see Figure 4a. Nevertheless, the slight downward shift of the center-of-mass position for $R/l \approx 0.4\text{--}0.5$ in Figure 4b is also correctly reproduced for electrodes of both short and intermediate length.

As an alternative to the full numerical calculations, we can also evaluate the energy landscapes by approximating the electrostatic energy using the simple geometric approximation proposed by 't Mannetje et al.^[30,41] This analytical calculation involves approximating the condensate drop-dielectric system as an electrical circuit consisting of two parallel plate capacitors in series formed by the overlap between the conducting drop and the electrodes. The overall capacitance of the system is approximated as $C(x, y) = \epsilon_0 \epsilon_d / d \cdot A_{\text{cap}}$, where $A_{\text{cap}} = A_1 A_2 / (A_1 + A_2)$, and A_1 and A_2 are the spatially varying overlap areas between the drop footprint and the two electrodes (see inset in Figure 6). The associated electrostatic energy of the system

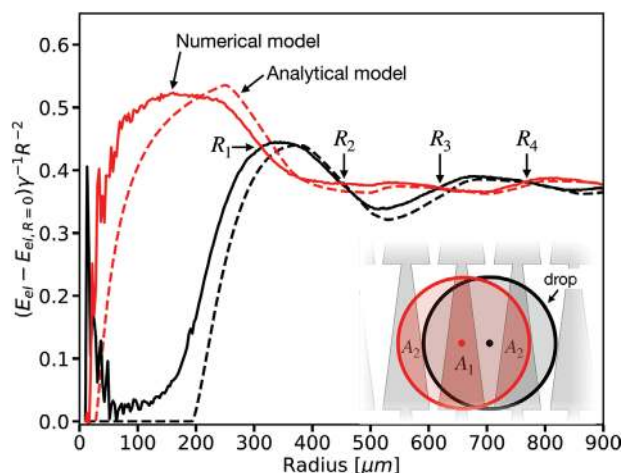


Figure 6. Normalized excess electrostatic energy versus drop size for drops centered on the gap (black) versus centered on the electrode (red). Solid: full numerical model; dashed: analytical model based on drop-electrode overlap areas A_1 , A_2 . R_1 , ... R_4 : critical radii of transitions from preferential gap center to electrode center-alignment. Inset: illustration of competing drop positions.

on application of an electrical voltage (U) can be written as $E_{el, cap} = -C(x, y)U^2/2$.^[41] The dashed lines in Figure 6 show the electrostatic energy minimum in this approximation for a drop centered on the gap (black) and on the electrode (red). Like in the case of the full numerical model, for small drops (i.e., $R \lesssim 320 \mu\text{m}$), it is energetically more favorable for the drop to be centered on the gap, whereas for increasing R there is a succession of transitions between preferred alignment on the electrode center and the gap center. While the energies deviate substantially for the smallest drop sizes (for which the overlap area with one of the electrodes and hence the total energy can vanish), the agreement improves for increasing drop size, and the predictions for the various subsequent transitions of the drop positions (R_2 , R_3 , R_4) become remarkably good.

While some of the aspects described above are very specific to the present electrode configuration, the overall excellent agreement demonstrates the ability of the numerical model to reproduce the experiments, including even subtle aspects such as the transitions between various competing local minima of the overall energy landscape are correctly captured. For not too small droplets, the simple analytical model of geometric overlap also provides reasonable predictions between various competing drop configurations.

4. Discussion and Perspectives

The results presented here clearly demonstrate the flexibility of electric fields in controlling condensation patterns on solid surfaces with submerged co-planar electrodes. While individual drops are obviously subject to their specific local environment, averaging over large ensembles shows that condensed drops accumulate at the local minima of electrostatic energy landscapes of remarkable complexity. Consequently, the drops undergo gradual translations as well as discrete transitions as local minima shift or become unstable with increasing

drop size. Apparently, the random character of coalescence events with neighboring drops, in combination with enhanced mobility of drops caused by the reduced contact angle hysteresis in EW with AC voltage,^[34] provide sufficient energy for the drops to explore the entire energy landscape despite the fact that energetic barriers between adjacent minima are obviously substantially larger than thermal energies. While not exploited here explicitly, compared to passive chemical or topographic patterning, EW-functionalization offers the advantage of switchability in addition to the enhanced mobility thanks to the reduced effective hysteresis.

While drop positions are well-defined and controllable beyond a certain critical size, the random distribution of small drops in Figure 3a confirms the earlier observation that the nucleation sites for drop condensation seem to be unaffected: no correlation can be observed between the positions of the smallest drops and the location of the electrodes. This arises from the fact that the forming liquid nuclei only experience a dielectrophoretic polarization force. Upon nucleation, this electrostatic force competes with surface forces caused by random heterogeneities on the surface. While the electric force scales with the (very small) volume of the critical nucleus, that is, $\propto R^3$, the latter scale with surface area, that is, $\propto R^2$ and therefore dominate. A control of nucleation rates and locations is therefore only possible if local electric fields and field gradients can be substantially increased, for example, by generating miniaturized electrode patterns on the nanoscale.

From an applied perspective, the key question for both fog harvesting and enhanced heat transfer is how the removal of drops from the surface can be optimized to condense as much liquid as possible. Obviously, this requires a somewhat broader perspective of the entire system than only the control of drop distribution patterns. While the results presented above clearly show that suitable electrode patterns allow to control drop positions and to promote faster growth by inducing lateral and vertical translations and coalescence, the same strong electrostatic forces also generate deep energetic traps that can hold back even large drops as shown in Figure 2 and thereby hamper efficient drop removal. To circumvent this problem, the electrically induced wettability patterns should be applied in combination with some form of time-dependent actuation. The easiest approach is to periodically activate the electrodes to induce drop motion and growth, and to subsequently deactivate them such that drops larger than the critical shedding radius can spontaneously shed off the surface under the influence of gravity. While some success of this strategy has been demonstrated,^[40,43] the overall performance was not impressive. In part, this is probably caused by the fact that the pinning forces increase as the EW-induced reduction of the contact angle hysteresis ceases upon switching off the AC voltage and hence the critical shedding radius increases and the shedding frequency decreases.^[44] Alternatively, one could make use of active transport strategies borrowed from EW-based lab-on-a-chip systems, where drops are transported toward activated electrodes.^[28] Given the nature of drop condensation, it is obviously not desirable to bring the condensing drops in direct contact with electrodes on top of the functionalized surface. Therefore, structured electrodes, possibly in two layers, should be embedded into the substrate and actuated in such a manner that they lead to a conveyor

belt-like directed motion. Such strategies are rather straightforward to implement for surfaces that are flat or covered by some “moderate” degree of topographic pattern. For intrinsically 3D structures such as meshes that are frequently used for fog harvesting the implementation of any form EW-enhanced condensation and drop removal is much more difficult to realize—notwithstanding initial demonstrations with crossing fibers of switchable wettability.^[45]

While the effect of EW on the drop distribution patterns is rather striking, the reported consequences for the total condensation rate and the resulting heat transfer are far less impressive.^[33,37,40] Applying standard models of dropwise heat transfer,^[46] Wikramanayake et al. pointed out that the majority of previous EW experiments were not very enlightening because they were carried out using water vapor in moist air.^[37] Under such conditions, it is well-known in the heat transfer community that the overall heat transfer resistance is dominated by the ambient air, which acts as a non-condensable background gas and introduces a diffusive boundary layer at the solid-vapor interface.^[47] The expected beneficial effects of EW-enhanced drop removal on heat transfer are thus overshadowed by mass transport limitations across that boundary layer and thus the actual potential of the EW-induced enhancement does not become evident. To demonstrate and exploit the benefits of EW, condensation setups should thus be designed in such a manner that drop removal is indeed the dominating factor for the overall heat transfer coefficient. This implies in particular preferential operation in pure vapor.

A final essential issue for any practical application of the effects described above is the stability of the EW-functionalized surfaces over extended periods of time. Like many other applications, both fog collection and heat transfer require long continuous operation times of the devices, ideally of the order of years. While proof-of-principle experiments in laboratories on short time scales are often relatively easy to achieve, maintaining cleanliness and hydrophobicity of coatings in the presence of complex and reactive fluids such as condensing water vapor is extremely demanding from a materials perspective. Recent experiments demonstrated that fluoropolymer surfaces commonly used in EW spontaneous charge up upon contact with water for several hours.^[48] In the presence of electric fields, this effect is even more pronounced and can even be exploited to generate well-controlled charge densities and charge patterns.^[49,50] Therefore, the development of reliable hydrophobic fluoropolymer coatings that remain stable throughout the life time of various types of devices has been a long standing challenge in applied EW research. One recommendation from these investigations has been to avoid water as an operating fluid whenever possible.^[51] While this is an interesting option for heat transfer devices, it is obviously not possible for fog harvesting applications. In such cases, novel materials with improved stability^[52] will be required to achieve the necessary stability of operation. Nevertheless, it is also worth pointing out that extended EW-enhanced drop condensation tests over a period of 40 h displayed - after some initial degradation within the first 1–2 h - rather stable operation even for conventional fluoropolymer surfaces (Figure S5, Supporting Information).

5. Conclusions

Co-planar electrodes embedded into electrowetting-functionalized surfaces allow to control the distribution of drops of condensing water vapor. For interdigitated electrodes with zigzag-shaped edges drops undergo a series of transitions between different preferred locations as they grow in size upon further condensation and coalescence. Comparison to numerical calculations shows excellent agreement with the experiments and demonstrates that the drops on average align at the minima of the drop size-dependent electrostatic energy landscape, including subtle transitions between preferred locations. This agreement demonstrates that the existing numerical approach provides a solid basis for future more sophisticated models that will include time-dependent electrical actuation schemes. Such models can eventually be used by engineers for numerical optimization of electrode designs and operation modes of future EW-enhanced drop condensation systems. A critical assessment of bottlenecks for such applications indicates that reducing mass transfer limitations and - in particular - the development of combinations of long term stable condenser surface materials and fluids will be essential for the technological success of the approach.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

dropwise condensation, electrowetting, fog harvesting, heat transfer, wetting

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- [1] D. Beysens, *Atmos. Res.* **1995**, 39, 215.
- [2] A. R. Parker, C. R. Lawrence, *Nature* **2001**, 414, 33.
- [3] K. C. Park, P. Kim, A. Grinthal, N. He, D. Fox, J. C. Weaver, J. Aizenberg, *Nature* **2016**, 531, 78.
- [4] C. R. Tracy, N. Laurence, K. A. Christian, *Am. Nat.* **2011**, 178, 553.
- [5] F. T. Malik, R. M. Clement, D. T. Gethin, W. Krawszik, A. R. Parker, *Bioinspiration Biomimetics* **2014**, 9, 3.

- [6] D. Milani, A. Abbas, A. Vassallo, M. Chiesa, D. A. Bakri, *Chem. Eng. Sci.* **2011**, *66*, 2491.
- [7] A. Lee, M. W. Moon, H. Lim, W. D. Kim, H. Y. Kim, *Langmuir* **2012**, *28*, 10183.
- [8] S. Wu, D. Meng, C. Chen, L. A. Shi, L. Zhou, Z. Huang, J. Li, Y. Hu, D. Wu, *Adv. Mater. Interfaces* **2019**, *6*, 1.
- [9] A. D. Khawaji, I. K. Kutubkhanah, J. M. Wie, *Desalination* **2008**, *221*, 47.
- [10] J. M. Beér, *Prog. Energy Combust. Sci.* **2007**, *33*, 107.
- [11] J. R. Barbosa, G. B. Ribeiro, P. A. De Oliveira, *Heat Transfer Eng.* **2012**, *33*, 356.
- [12] M. H. Kim, C. W. Bullard, *Int. J. Refrig.* **2002**, *25*, 924.
- [13] J. W. Rose, *Proc. Inst. Mech. Eng., Part A* **2002**, *216*, 115.
- [14] Y. Hou, M. Yu, X. Chen, Z. Wang, S. Yao, *ACS Nano* **2015**, *9*, 71.
- [15] B. Mondal, M. Mac Giolla Eain, Q. F. Xu, V. M. Egan, J. Punch, A. M. Lyons, *ACS Appl. Mater. Interfaces* **2015**, *7*, 23575.
- [16] A. Ghosh, S. Beaini, B. J. Zhang, R. Ganguly, C. M. Megaridis, *Langmuir* **2014**, *30*, 13103.
- [17] R. D. Narhe, D. A. Beysens, *Phys. Rev. Lett.* **2004**, *93*, 076103.
- [18] J. B. Boreyko, C. H. Chen, *Phys. Rev. Lett.* **2009**, *103*, 2.
- [19] N. Miljkovic, R. Enright, E. N. Wang, *ACS Nano* **2012**, *6*, 1776.
- [20] N. Miljkovic, D. J. Preston, R. Enright, E. N. Wang, *ACS Nano* **2013**, *7*, 11043.
- [21] P. B. Weisensee, Y. Wang, H. Qiang, D. Schultz, W. P. King, N. Miljkovic, *Int. J. Heat Mass Transfer* **2017**, *109*, 187.
- [22] S. Anand, A. T. Paxson, R. Dhiman, J. D. Smith, K. K. Varanasi, *ACS Nano* **2012**, *6*, 10122.
- [23] H. Tsuchiya, M. Tenjimbayashi, T. Moriya, R. Yoshikawa, K. Sasaki, R. Togasawa, T. Yamazaki, K. Manabe, S. Shiratori, *Langmuir* **2017**, *33*, 8950.
- [24] R. De Ruiter, C. Semperebon, M. Van Gorcum, M. H. Duits, M. Brinkmann, F. Mugele, *Phys. Rev. Lett.* **2015**, *114*, 234501.
- [25] M. G. Pollack, R. B. Fair, A. D. Shenderov, *Appl. Phys. Lett.* **2000**, *77*, 1725.
- [26] S. K. Cho, H. Moon, C. J. Kim, *J. Microelectromech. Syst.* **2003**, *12*, 70.
- [27] F. Mugele, J.-C. Baret, *J. Phys.: Condens. Matter* **2005**, *17*, R705.
- [28] F. Mugele, J. Heikenfeld, *Electrowetting: Fundamental Principles and Practical Applications*, Wiley-VCH, Weinheim, **2019**.
- [29] U. C. Yi, C. J. Kim, *J. Micromech. Microeng.* **2006**, *16*, 2053.
- [30] D. 't Mannetje, S. Ghosh, R. Lagraauw, S. Otten, A. Pit, C. Berendsen, J. Zeegers, D. Van Den Ende, F. Mugele, *Nat. Commun.* **2014**, *5*, 3559.
- [31] R. De Ruiter, A. M. Pit, V. M. De Oliveira, M. H. Duits, D. Van Den Ende, F. Mugele, *Lab Chip* **2014**, *14*, 883.
- [32] J. Kim, M. Kaviany, *J. Appl. Phys.* **2007**, *101*, 10.
- [33] D. Baratian, R. Dey, H. Hoek, D. Van Den Ende, F. Mugele, *Phys. Rev. Lett.* **2018**, *120*, 214502.
- [34] F. Li, F. Mugele, *Appl. Phys. Lett.* **2008**, *92*, 244108.
- [35] R. Yan, T. S. McClure, I. H. Jasim, A. K. R. Koppula, S. Wang, M. Almasri, C. L. Chen, *Appl. Phys. Lett.* **2018**, *113*, 204101.
- [36] R. Yan, C.-L. Chen, *J. Heat Transfer* **2019**, *141*, 111501.
- [37] E. D. Wikramanayake, V. Bahadur, *Int. J. Heat Mass Transfer* **2019**, *140*, 260.
- [38] E. D. Wikramanayake, J. Perry, V. Bahadur, *Appl. Phys. Lett.* **2020**, *116*, 193701.
- [39] S. Högnadóttir, K. Kristinsson, H. G. Thormar, K. Leosson, *Appl. Phys. Lett.* **2020**.
- [40] R. Dey, J. Gilbers, D. Baratian, H. Hoek, D. van den Ende, F. Mugele, *Appl. Phys. Lett.* **2018**, *113*, 243703.
- [41] D. Mannetje, A. Banpurkar, H. Koppelman, M. H. Duits, D. Van Den Ende, F. Mugele, *Langmuir* **2013**, *29*, 9944.
- [42] A. Cavalli, M. Musterd, F. Mugele, *Phys. Rev. E Stat., Nonlinear, Soft Matter Phys.* **2015**, *91*, 023013.
- [43] E. Wikramanayake, V. Bahadur, *Colloids Surf. A* **2020**, *599*, 124874.
- [44] T. D. Mannetje, C. U. Murade, D. Van Den Ende, F. Mugele, *Appl. Phys. Lett.* **2011**, *98*, 014102.
- [45] H. B. Eral, J. De Ruiter, R. De Ruiter, J. M. Oh, C. Semperebon, M. Brinkmann, F. Mugele, *Soft Matter* **2011**, *7*, 5138.
- [46] Y. Zhao, D. J. Preston, Z. Lu, L. Zhang, J. Queeney, E. N. Wang, *Int. J. Heat Mass Transfer* **2018**, *119*, 931.
- [47] W. Minkowycz, E. Sparrow, *Int. J. Heat Mass Transfer* **1966**, *9*, 1125.
- [48] A. G. Banpurkar, Y. Sawane, S. M. Wadhai, C. U. Murade, I. Siretanu, D. Van Den Ende, F. Mugele, *Faraday Discuss.* **2017**, *199*, 29.
- [49] H. Wu, R. Dey, I. Siretanu, D. van den Ende, L. Shui, G. Zhou, F. Mugele, *Small* **2020**, *16*, 2.
- [50] H. Wu, N. Mendel, S. van der Ham, L. Shui, G. Zhou, F. Mugele, *Adv. Mater.* **2020**, *32*, 2001699.
- [51] B. Raj, M. Dhindsa, N. R. Smith, R. Laughlin, J. Heikenfeld, *Langmuir* **2009**, *25*, 12387.
- [52] A. T. Paxson, J. L. Yagüe, K. K. Gleason, K. K. Varanasi, *Adv. Mater.* **2014**, *26*, 418.