

How Spontaneous Electrowetting and Surface Charge Affect Drop Motion

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Water drops sliding on hydrophobic surfaces spontaneously separate charges at their rear. It is unclear how this charge separation affects the contact angles of a sliding drop. We slide grounded and insulated drops on hydrophobic surfaces at low capillary numbers ($\leq 10^{-4}$). We find that the drop charge leads to spontaneous electrowetting that decreases the contact angles. The deposited charges lead to a surface charge effect and decrease the contact angle. Both phenomena compensate each other at the receding contact line, resulting in an insignificant change in the receding contact angle of a sliding drop.

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Introduction—Drop sliding is a universal phenomenon and spans multiple disciplines. For instance, the back surface of a desert beetle inspires research into the development of water harvesting technologies [1–3], dropwise condensation for enhanced heat transfer [4], retention of agricultural sprays on plant leaves [5], and drug production using droplet microfluidics [6]. To achieve the desired wetting properties, the surface energy is lowered by employing a water repellent coating. When a water drop slides on a hydrophobized surface, charges separate at the receding contact line, and the drop acquires a net charge [7–14]. This phenomenon is called “slide electrification or solid-liquid contact electrification” [9,10,13,15]. Slide electrification alters the governing forces on the drop and influences drop motion [16]. By sliding grounded and insulated drops at low capillary numbers, we investigate the influence of slide electrification on the sliding contact angles.

Slide electrification of drops can lead to two phenomena, which decrease the contact angles (CAs) of a sliding drop. The first is electrowetting. The drop acquires a net positive charge, which originates from the negative charges left behind on the surface. This net positive charge in the drop causes an electric potential U with respect to the

ground electrode beneath the substrate [17,18]. Integrating the energy of the electric field across the thickness (d) of the dielectric material and assuming that the drop size is much larger than d leads to an energy per unit area of $\epsilon_o \epsilon_s U^2 / 2d$ [19–22]. Formally, this can be viewed as a reduction in the solid-liquid interfacial energy ($\Delta\gamma_{SL}$) [red arrows, Fig. 1(a)]. It can also be interpreted as an electromechanical force per unit contact line length generated by a nonuniform electric field [Fig. 1(a)] [22]. A decrease in solid-liquid interface energy in turn decreases the advancing contact angle (θ_a) at the front and receding contact angle (θ_r) at the rear of the sliding drop. The decrease in θ_a and θ_r is described by

$$\cos \theta_{a/r}(U) - \cos \theta_{a/r}(U=0) = -\frac{\Delta\gamma_{SL}}{\gamma} = \frac{\epsilon_o \epsilon_s}{2d} \cdot \frac{U^2}{\gamma} \quad (1)$$

Here, $\theta_{a/r}(U=0)$ and $\theta_{a/r}(U)$ are the advancing or receding CAs in the absence and presence of a drop potential U , respectively, γ is the surface tension of the liquid, ϵ_s and ϵ_o are the dielectric permittivities of the substrate and vacuum, respectively. In typical electrowetting experiments, U is applied to the drop by an external electrode. However, in slide electrification, the charges separate spontaneously during the drop motion. Hereafter, we term this phenomenon “spontaneous electrowetting.” The negative charge density at the solid-liquid interface (not shown in Fig. 1), which forms spontaneously when water contacts a hydrophobic solid surface, is compensated within the Debye layer by the acquired positive counterions in the drop. The contribution of the surface charge in the Debye layer does not appear in Eq. (1): its effect can be

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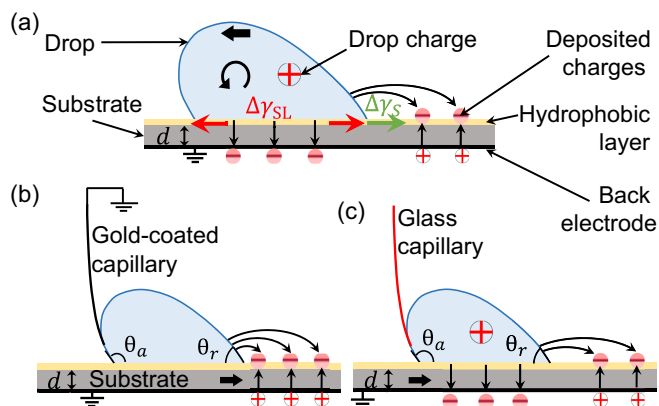


FIG. 1. (a) A drop sliding on a hydrophobic surface is subjected to spontaneous electrowetting and surface charge effects. Drop charge results in spontaneous electrowetting ($\Delta\gamma_{SL}$) and deposited charges increase solid surface energy behind the drop ($\Delta\gamma_S$). Both effects reduce the CAs of a sliding drop and coexist with viscous dissipation (circular arrow). The horizontal black arrow in (a) indicates the direction of drop motion. The vertical and curved black arrows indicate the electric field lines. The thickness of the hydrophobic layer is $\sim\text{nm}$ (yellow) and the substrate is $\sim\text{mm}$ (gray). (b) Drop sliding using a gold-coated electrically conductive capillary. It constantly discharges the drop and maintains it at zero electric potential. (c) Drop sliding using a glass capillary. It results in charges staying in the drop. The black horizontal arrows in substrates of (b),(c) indicate the direction of stage motion.

neglected because the related surface potential is much lower than the drop potential U .

The second possible phenomenon in slide electrification is the change in wettability of the surface due to the deposition of charges onto the surface [23,24]. Depositing charges onto the surface requires electrostatic work to be carried out, which effectively increases the solid-air interfacial energy ($\Delta\gamma_S$) [green arrow, Fig. 1(a)]. It can also be interpreted as an electromechanical force per unit receding contact line length generated by a nonuniform electric field [22]. This $\Delta\gamma_S$ scales quadratically with the surface charge density on the surface [Eq. (2)] [23]. This increase in surface energy decreases the CA of the drop. Using Young's equation locally at the rear contact line yields

$$\cos \theta_r(\sigma) - \cos \theta_r(\sigma = 0) = \frac{\Delta \gamma_S}{\gamma} \propto \frac{\sigma^2}{\gamma} \quad (2)$$

Here, $\theta_r(\sigma)$ is the receding CA with charge separation, $\theta_r(\sigma = 0)$ is the CA without charge separation, and σ is the surface charge density at the solid-air interface. The precise value of $\Delta\gamma_S$ depends on the shape and size of the charged region. We term this phenomenon the “surface charge effect.”

Both effects, spontaneous electrowetting and surface charge, depend on the charge separation. Charge separation depends on the drop velocity v [25,26] and surface wetting

properties [27,28]. Furthermore, they coexist with hydrodynamic viscous dissipation [29,30]. Viscous dissipation inside the drop increases the advancing CA and decreases the receding CA of a sliding drop at high capillary numbers [31–33], $Ca = (\mu v / \gamma) > 10^{-3}$. Here, μ is the dynamic viscosity of the liquid.

All three phenomena—spontaneous electrowetting, surface charge, and viscous dissipation—affect the contact angles and determine the forces acting on the sliding drop. However, it is unclear to what extent slide electrification contributes to the contact angles of a sliding drop. In this Letter, we separate and quantify spontaneous electrowetting and surface charge effects in the absence of viscous dissipation. Both effects need to be considered for a complete understanding of the physics of drop motion.

In contrast to tilted plate experiments [9,28,34,35], we mitigate viscous dissipation by sliding 5 μL milli-Q water drops at constant velocities in the range 0.1–10 mm/s ($Ca \sim 10^{-6} - 10^{-4}$) [36–39]. Sliding a drop using an electrically conductive capillary results in a grounded drop, i.e., the drop slides at zero electric potential [Fig. 1(b)]. This allows us to omit spontaneous electrowetting [40]. As a result, only the deposited charges behind the drop affect the drop sliding motion, allowing us to investigate the surface charge effect. In contrast to a conductive capillary, we use a glass capillary for a sliding drop to investigate the combined effects of spontaneous electrowetting and surface charges [Fig. 1(c)].

Methods—We coat Trichloro(1H,1H,2H,2H-perfluorooctyl)silane (PFOTS, 97% Alfa Aesar) and Trichloro(octyl)silane (OTS, Sigma Aldrich 97%) on $76.2 \times 25.4 \times 1$ (or 0.25, 0.5, 2, 5) mm³ fused quartz (Thermo Fisher Scientific, resistivity $\approx 10^{15}$ Ω cm, $\epsilon_s = 3.5$) using a chemical vapor deposition process [41] [supplement Material (SM) Sec. 1]. For simplicity, the fused quartz substrate will be termed “quartz” hereafter. We characterize the surfaces using surface energy measurements (SM Sec. 2), scanning force microscopy (SM Sec. 3), and the sessile drop method (SM Sec. 4). We measure sliding CAs for 5 μ L milli-Q water drops (Sartorius, resistivity ≈ 18.2 M Ω cm, $\gamma = 72$ mN/m). In all measurements, we slide drops for 5 cm. The substrates are placed on a metal plate, which is connected to ground. The CAs are recorded by a side-view camera [36–40,42]. To determine advancing (leading) and receding (trailing) CAs of the drops, a tangent is fit to 10 pixels (image resolution = 6 μ m/pixel) above the baseline [43,44]. To make a capillary conductive, the glass capillary is sputter-coated with 5 nm thick chromium followed by a 30 nm thick gold layer (SM Sec. 5).

Results—Sliding a grounded drop deposits charges at zero potential; in parallel countercharges in the grounded drop are continuously neutralized. The deposited charges at the surface increase the solid surface energy ($\Delta\gamma_S$). For a water drop sliding at 2 mm/s on a neutral (or unwetted)

PFOTS quartz, we measure advancing and receding CAs of $120^\circ \pm 2^\circ$ and $60^\circ \pm 3^\circ$, respectively. Both CAs stay constant after a sliding length ≈ 4 mm and do not change in the kinetic regime of drop sliding [Fig. 2(a), black and gray curves]. For an electrically insulated 5 μL water drop sliding on the same neutral PFOTS quartz, we observe that the advancing CA decreases to 113° after 15 mm stage motion. Thereafter, it remains constant at $113^\circ \pm 2^\circ$ along the drop sliding path [Fig. 2(a), red and pink curves]. We attribute the decrease in advancing CA to the charging of the sliding drop. As the drop starts sliding, it deposits charges, leading to a surface charge density $\sigma(x)$ at the dewetted surface. Here, x is a coordinate along the drop path. The charge in the drop is equal in magnitude but opposite in polarity to the deposited charges. The accumulated charges in the drop develop an electric potential U , which increases according to

$$U = -\frac{w}{C} \int_0^L \sigma dx \quad (3)$$

Here, w is the width of the drop, C is its capacitance, and L is the slide length. As U increases, fewer and fewer charges are deposited at the surface. After a slide length of ≈ 15 mm, the drop saturates with charges, and no countercharges are deposited at the dewetted surface. This length is a direct measurement of the saturation length (λ), which is not directly measurable in tilted plate experiments [9,13]. This value agrees with estimated saturation lengths in the literature [18].

Once the drop saturates with charges, we measure a difference of 7° between the advancing CA of the grounded and charge-saturated drops [Fig. 2(a)]. Since there are no charges on the PFOTS quartz in front of the drop, the decrease in advancing CA of an insulated drop is due to spontaneous electrowetting [left side, Fig. 2(b)]. Using Eq. (1) we calculate a drop potential of ≈ 700 V. It is of the same order as reported for drops sliding down a tilted plane [8,17,18]. This spontaneous drop potential corresponds to a decrease of 8 ± 3 mN/m in solid-liquid interfacial energy ($\Delta\gamma_{\text{SL}}$). The error represents variation along the drop path.

For the same insulated drop, we determine that the receding CA decreases to 60° at the onset of sliding and remains at 60° in the kinetic regime of drop sliding [Fig. 2(a), red curve]. In particular, the receding CA for both grounded and insulated drops is similar. Because of the increasing drop potential and corresponding electrowetting effect, one would expect a lower receding CA for an insulated drop compared to a grounded drop. But why is the receding CA of both the grounded and insulated drops the same? Unlike at the advancing contact line, where only spontaneous electrowetting occurs, the receding contact line is subject to two simultaneous phenomena: spontaneous electrowetting and the surface charge effect. At the onset of drop motion, the drop deposits charges at zero

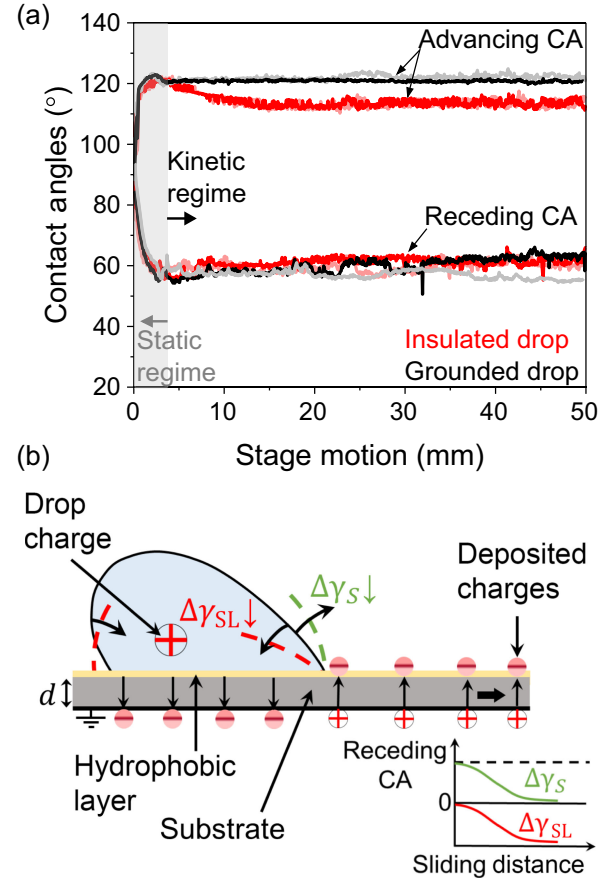


FIG. 2. (a) CAs for grounded and insulated drops that slide on a PFOTS-quartz surface. The thickness of quartz is 1 mm. The grounded drop is sliding at zero potential, while the insulated drop accumulates charges. The black and gray data correspond to two independent measurements for grounded drops and the red and pink correspond to two independent measurements for insulated drops of 5 μL milli-Q water drops sliding at 2 mm/s. (b) The advancing CA of an insulated drop decreases due to spontaneous electrowetting, sketched as the change in black drop profile to the red dashed line. The receding CA of an insulated drop is subjected to two coupled phenomena: spontaneous electrowetting and the surface charge effect. Spontaneous electrowetting decreases the receding CA, sketched as the change in black drop profile to red dashed line. Simultaneously, the decrease in surface charge effect increases the receding CA, sketched as the change in black line to green dashed line. The receding CA of a grounded drop is only subjected to the surface charge effect. The black horizontal arrow indicates the direction of stage motion. Vertical arrows in the substrate are electric field lines. The inset schematic shows a qualitative change in solid-air interfacial energy ($\Delta\gamma_{\text{S}}$) that is compensated by the change in solid-liquid interfacial energy ($\Delta\gamma_{\text{SL}}$). This compensation results in an insignificant change in receding CA (black dashed line).

potential and increases the solid surface energy ($\Delta\gamma_{\text{S}}$). As soon as the drop begins sliding, it accumulates the opposite charges. These charges in the drop develop a potential U . This drop potential decreases the charge separation at the receding contact line. With further contact line motion, the

deposited surface charge density decreases to $\sigma \cong 0$ and the drop charge saturates. According to Eq. (2) ($\Delta\gamma_S \sim \sigma^2$), a decrease in σ leads to a decrease in $\Delta\gamma_S$, which increases the receding CA [green dashed line at receding CA, Fig. 2(b)]. At the same time, the drop potential decreases the solid-liquid interface energy according to Eq. (1) ($-\Delta\gamma_{SL} \sim U^2$), which leads to a decrease in the receding CA [red dashed line at receding CA, Fig. 2(b)]. This decrease in potential scales with the surface charge density σ [Eq. (3)]. Therefore, according to Eq. (1), $-\Delta\gamma_{SL} \sim \sigma^2$. We find that $\Delta\gamma_{SL}$ for the ungrounded drop is equal to the $\Delta\gamma_S$ for the grounded drop: $\Delta\gamma_{SL} = \Delta\gamma_S \propto \sigma^2$. Both effects, spontaneous electrowetting and surface charge, compensate each other at the receding contact line during the entire drop motion [inset Fig. 2(b)]. As a result, the receding CA of $60^\circ \pm 3^\circ$ for an insulated drop resembles that of a grounded drop [Fig. 2(a)].

In typical electrowetting experiments on dielectrics, Nelson *et al.* found that external potential leads to a decrease in the advancing and receding CAs of a sliding drop at capillary numbers $10^{-5} \leq Ca \leq 10^{-3}$ [45]. It does not alter CA hysteresis ($CAH = \theta_a - \theta_r$) [45]. However, in the kinetic regime of drop sliding at a $Ca \sim 10^{-5}$, we find that the CAH ($\theta_a - \theta_r$) reduces by 7° for an insulated drop [Fig. 2(a)]. The reason for the decrease in CAH is due to a decrease in the advancing CA by spontaneous electrowetting and an insignificant change in the receding CA [Fig. 2(b)].

We develop a mathematical model to estimate the surface charge effect ($\Delta\gamma_S$) on the dewetted surface (SM Sec. 9). We assume a one-dimensional insulating layer of thickness d (which extends infinitely in the other two directions) and dielectric permittivity ϵ_s that is placed on a grounded plate. For such a system, the change in solid surface energy is

$$\Delta\gamma_S = \frac{\sigma^2 d}{2\epsilon_0 \epsilon_s} \quad (4)$$

The insulator-air interface is charged with a uniform surface charge density σ . An additional grounded plate is placed in air at a large distance $L \gg d$. Equation (4) effectively models the effect of the Maxwell stress tensor acting on the liquid-air interface. Taking a quartz thickness of $d = 0.25$ mm, a relative permittivity $\epsilon_s = 3.5$, and a surface charge density of $\sigma \approx 60 \mu\text{C}/\text{m}^2$, which is measured by Kelvin probe [SM Fig. S5(b)] in Eq. (4) provides an increase in surface energy ($\Delta\gamma_S$) of $15 \text{ mN}/\text{m}$. Therefore, our model [Eq. (4)] provides a good estimation for $\Delta\gamma_S$ for thin insulating substrates with a thickness smaller than the drop size.

Influence of substrate thickness: We systematically vary the thickness of the PFOTS-quartz substrate as 0.25, 0.5, 1, 2, and 5 mm. For $5 \mu\text{L}$ drops sliding at $2 \text{ mm}/\text{s}$ speed, we measure identical receding CAs for both grounded and insulated drops on the respective quartz

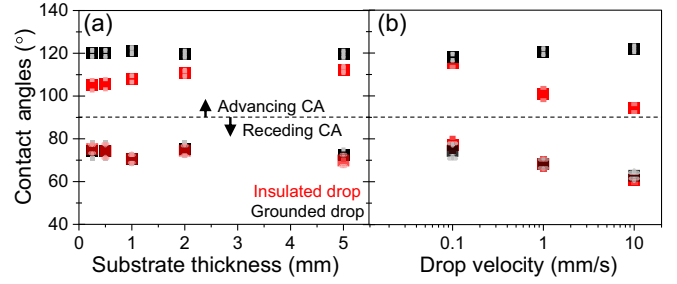


FIG. 3. (a) Average CAs for grounded and insulated $5 \mu\text{L}$ milli-Q water drops sliding at $2 \text{ mm}/\text{s}$ on PFOTS quartz with quartz thicknesses 0.25, 0.5, 1, 2, and 5 mm. (b) Average CAs for $5 \mu\text{L}$ water drops sliding at drop velocities of 0.1, 1, and $10 \text{ mm}/\text{s}$ on 1 mm thick PFOTS quartz. We took values after a stage motion of 3–5 cm (kinetic regime) for determining the average. Error bars correspond to variation in CAs for three independent drop sliding measurements.

thicknesses [Fig. 3(a)]. Surprisingly, we also measure identical receding CAs for grounded and insulated drops on substrate thicknesses of 2 and 5 mm. For these two thicknesses, we anticipate that the parallel plate capacitor models employed in Eqs. (1) and (4) are not applicable since the drop size is comparable to the substrate thicknesses. Our results imply that spontaneous electrowetting and the surface charge effect compensate each other at the receding contact line for the quartz thicknesses in the range 0.25–5 mm.

The difference between the advancing CA of the grounded and insulated drops decreases with the increasing thickness of the quartz [Fig. 3(a)]. Using Eq. (1), we calculate the maximum decrease in $\Delta\gamma_{SL} \approx 17 \text{ mN}/\text{m}$ for the 0.25 and 0.5 mm thicknesses.

Influence of drop velocity: We slide $5 \mu\text{L}$ water drops at 0.1, 1, $10 \text{ mm}/\text{s}$ on 1 mm thick PFOTS quartz. This velocity range corresponds to capillary numbers $10^{-6} \leq Ca \leq 10^{-4}$ for a water drop. We measure identical receding CAs for both the grounded and insulated drops for the respective drop velocities [Fig. 3(b)]. Thus, spontaneous electrowetting and surface charge compensate at the receding contact line irrespective of the drop velocity.

For grounded drops, we observe a slight increase in advancing CA and a decrease in receding CA with increasing drop velocity [black data, Fig. 3(b)]. Since the charge separation for a grounded drop does not depend on drop velocity in this range for silanized surfaces [40], we attribute this observation to dissipation of the moving contact line described by the molecular kinetic theory [46,47]. Contrary to grounded drops, the advancing CA of insulated drops decreases with increasing drop velocity [Fig. 3(b)]. We attribute this decrease in advancing CA to an increasing electric potential in the drop. With an increase in potential, the strength of spontaneous electrowetting increases [Eq. (1)]. At lower velocity, drops may dissipate accumulated charges through the glass capillary or ambient, which results in a lower electric potential of the drop. Furthermore, we investigate the influence of salt by varying the NaCl concentration in water drops as 0.1, 1, 10, and

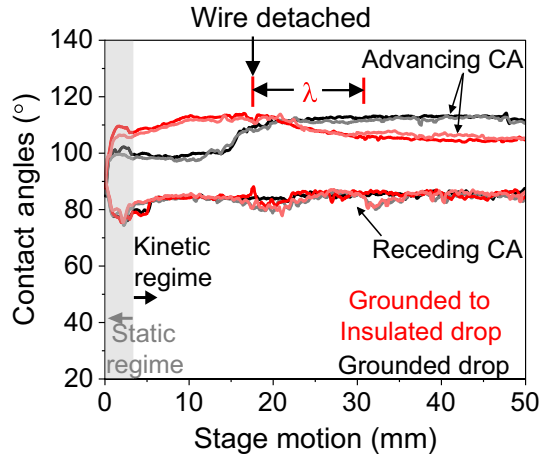


FIG. 4. CA variation of a sliding drop whose charging state is switched from grounded to an insulated state (red). Drop grounding is achieved by attaching a conductive wire of 0.2 mm diameter to the drop. Wire detachment from the sliding drop results in an insulated sliding drop. The instant of wire detachment is indicated by a vertical arrow. Immediately afterwards, a grounded drop is slid along the same path to detect the changes in CA due to surface charge density (black). The plotted pink CA data followed by gray data represent an independent repetition of the measurement. The data correspond to 5 μ L water drops sliding at 2 mm/s on a PFOTS coated 1 mm thick quartz substrate. The quartz thickness is 1 mm. The CA data are smoothed by a five-point moving average.

100 mM. For 5 μ L drops sliding at 2 mm/s, no significant difference is detected in the receding CA = $89^\circ \pm 5^\circ$ for grounded and insulated drops when the NaCl concentration is varied from 0.1 to 100 mM (SM Fig. S6).

We extend our experiments with a glass capillary having a flexible grounded wire (SM [48] Fig. S7). Sliding a drop with a wire attached to it results in drop sliding at zero potential. The detachment of the wire from the drop switches a grounded drop to an insulated drop while the drop is in motion. A grounded 5 μ L drop on a neutral PFOTS quartz results in advancing and receding CAs of 113° and 83° , respectively (Fig. 4). At about the 18 mm stage motion, we detach the wire from the drop and the drop accumulates charges. As a result, the advancing CA gradually decreases and stagnates at 105° from 32 mm of stage motion. The gradual decrease in the advancing CA takes place over the saturation length $\lambda = 14$ mm. This implies that the detachment of the grounded wire from the drop results in a decaying charge separation over the saturation length (λ) [Fig. 4; SM Fig. S7]. Once the drop is saturated with charges and reaches its maximum potential, the charge separation behind the drop vanishes ($\sigma = 0$).

To detect the surface charge density gradient formed by the previous drop, we immediately slide a second grounded 5 μ L drop along the same path. For the first 13.5 mm stage motion, we measure an advancing CA of 100° (Fig. 4). The reduction in the advancing CA is attributed to a surface charge effect induced by the preceding

grounded portion of the grounded-to-insulated sliding drop. Beyond 13.5 mm of stage motion, we observe a gradual increase in the advancing CA until 27 mm of stage motion. The stage position where the gradual increase in the advancing CA takes place is shifted by drop length, which in our case is 2.5 mm. Thereafter, the advancing CA remains constant at 113° for the rest of the stage motion. The gradual increase in the advancing CA of the grounded drop confirms the existence of a surface charge density gradient [also see SM Fig. S5(b)]. Regardless of the presence of a surface charge density gradient or drop charging state, we measure a constant receding CA of 83° . Therefore, spontaneous electrowetting and surface charge effects compensate each other ($\Delta\gamma_{SL} = \Delta\gamma_S$) at the receding CA during all stages of drop sliding.

Conclusion—The drop charge develops an electric potential ~ 1 kV and leads to spontaneous electrowetting. Deposited charges behind the drop lead to a surface charge effect and increase the surface-air interface energy ($\Delta\gamma_S$) ~ 10 mN/m. These two effects balance the receding CA irrespective of the drop charging state. Charge separation substantially influences the sliding contact angles, implying a significant contribution to the wetting dynamics of drops on surfaces.

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C.H. conceived the idea, designed the work, prepared the samples, performed the experiments, and analyzed the data. B.L. prepared and characterized the samples and performed the AFM experiments. C.H. and R.C. analyzed the contact angle data. N.K. performed Kelvin Probe experiments. A.D.R. suggested the principle of virtual work and noted the absence of viscous dissipation. S.S. and C.H. performed drop capacitance measurements. H.-J.B. proposed and derived the mathematical model. C.H., R.B., and H.-J.B. interpreted the data. C.H. and R.B. wrote the manuscript.

The authors declare no conflict of interest.

Data availability—The data that support the findings of this article are openly available at [85].

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