SUPPLEMENTARY INFORMATION

Temperature dependent electrowetting behavior on Teflon AF1600

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Dielectric Layer Characterisation

The contact angle CA in an electrowetting on dielectric (EWOD) setup is determined by the Young-Lippmann (YL) equation, which extends the force equilibrium at the triple line (Young-Dupré equation) with an expression derived from the electric field energy stored in the dielectric layer between the electrically conductive droplet and a bottom electrode. For thin and plane dielectric layers the energy can be calculated from the parallel plate capacitor $C = \epsilon_0 \cdot \epsilon_r \cdot A/d$ defined by the permittivity $\epsilon = \epsilon_0 \cdot \epsilon_r$, the thickness of the dielectric layer d, and the electrodes area A. The additional electric force at the triple line is calculated as $F_{\mathrm{el}} = c/2 \cdot U^2$, with the specific capacitance $c = C/A$. The necessary condition for the geometry $A \gg d^2$ is guaranteed by our sample and the droplet size, with d of $\sim$1 µm and A of $5 \sim 7$ mm^2. Additionally, a homogeneous, linear and isotropic dielectric material, characterised by a constant permittivity ϵ is required.

To determine the dielectric properties of our Teflon AF layer, we prepared a parallel plate capacitor setup by a thermally evaporated circular gold electrode ($D = 9$ mm) with a thickness of 150 nm on top of the Teflon AF surface.

The first characterisation was done by standard LCR-Meter measurements (Wayne Kerr WK6500). Standard LCR-meters evaluate sinusoidal AC signals to determine the electrical impedance of the sample, in a wide frequency range, but with limited amplitudes in the range of ~ 1 V. After standard open-short calibration of the measurement-setup, excitation voltage levels between 10 mV and 1 V at frequencies between 500 Hz and 100 kHz gave a capacitance $C = 1.021 \pm 0.001$ nF.

As we are specifically interested in the characterisation of our sample-layer in a wide voltage range up to 100 V at rather low frequencies, we chose a voltage versus current (IV) measurement method. From the definition of capacitance $C = Q/U$, as quotient of charge Q and voltage U, follows for the current $I = \dot{Q} = C \cdot \dot{U}$ for constant C (the dot denote the time derivative). An Agilent B2911A Precision Source/Measure Unit was used to supply a small constant current to our Teflon AF capacitor and measure the voltage at the sample.

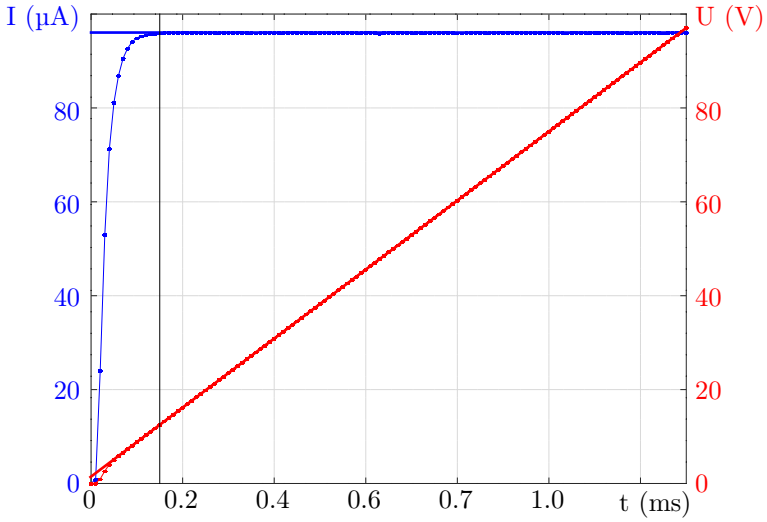

Figure S1: Typical IV measurement result. After a settling time of ≈0.15 ms the constant I and the constant $\dot{U} = \mathrm{d}U/\mathrm{d}t$ show the linear dielectric material with constant $C = I/\dot{U}$ up to a voltage of 100 V. The thick solid lines denote the linear fit.

We observed an perfect linear time-dependence of the measured voltage at various constant currents between −100 µA and 100 µA, proving the constant capacitance and dielectric constant of the Teflon AF layers. Figure S1 shows a typical IV result for $I = 96\,\mu\mathrm{A}$. After calibration of our measurement setup, the resulting capacity of the sample is constant and equal to the LCR results for voltages up to 100 V.

The specific capacitance of our Teflon layer is $c = C/A = 16.03\,\mu\mathrm{F/m^2}$.

Calculation of the Quasi-static Limit

As our investigations focus on the static description of the electrowetting process, we have to avoid any dynamic effects. The applied voltage leads to the CA and droplet shape changes, accordingly, the dynamic of the applied voltage has to be limited to justify the static analyses.

For small droplets, with a diameter below the capillary length ($d = 2.7$mm in the case of water), the shape of the droplet can be approximately calculated as simple spherical cap. The dimensions of the droplet are described using the spherical radius R and the total height H, the base line radius r and the CA θ, as shown in Fig. S2.

$$R = \frac{r}{\sin\theta} , \quad H = \frac{r}{\sin\theta} \cdot (1 - \cos\theta) . \tag{1}$$

The volume of the spherical cap is:

$$V = \frac{\pi}{3} \cdot H^2 \cdot (3 \cdot R - H) = \frac{\pi}{3} \cdot r^3 \cdot \frac{(2 + \cos\theta) \cdot (1 - \cos\theta)^2}{\sin^3\theta} , \tag{2}$$

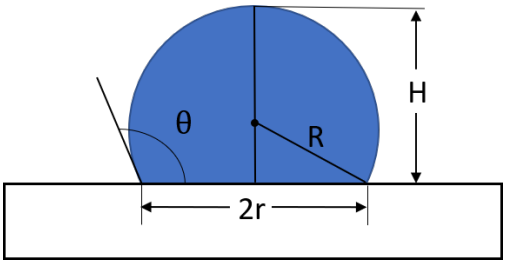

Figure S2: Sketch of a small droplet sitting on a hydrophobic surface, with CA $\theta > 90°$, droplet height H, droplet radius R and contact base radius r.

During the EWOD experiments the droplet volume is constant and the CA changes with voltage. Accordingly we write the droplet base-radius r in dependence of the CA θ:

$$r(\theta)^3 = \frac{3 \cdot V}{\pi} \cdot \frac{\sin^3\theta}{(1 - \cos\theta)^2 \cdot (2 + \cos\theta)} \tag{3}$$

The change of the CA with the applied voltage changes the base-circle diameter, causing the

movement of the triple-line:

$$\frac{\partial r}{\partial \theta} = \frac{-r(\theta)}{(2+\cos\theta)\cdot\sin\theta} \tag{4}$$

The variation of the CA θ with the applied voltage U is described by the Young-Lippmann equation:

$$\cos\theta = \cos\theta_Y + \frac{c\cdot U^2}{2\cdot\gamma_{\mathrm{la}}}\,, \tag{5}$$

$$\frac{\partial\theta}{\partial U} = \frac{-c\cdot U}{\gamma_{\mathrm{la}}\cdot\sin\theta(U)}\,. \tag{6}$$

with Young's CA θ_Y, the liquid-air interface tension γ_{la}, and the specific capacitance c of the dielectric layer. Finally we can calculate the speed $v = \partial r/\partial t$ of the triple-line, due to the applied voltage:

$$v = \frac{\partial r}{\partial\theta}\cdot\frac{\partial\theta}{\partial U}\cdot\frac{\partial U}{\partial t} \tag{7}$$

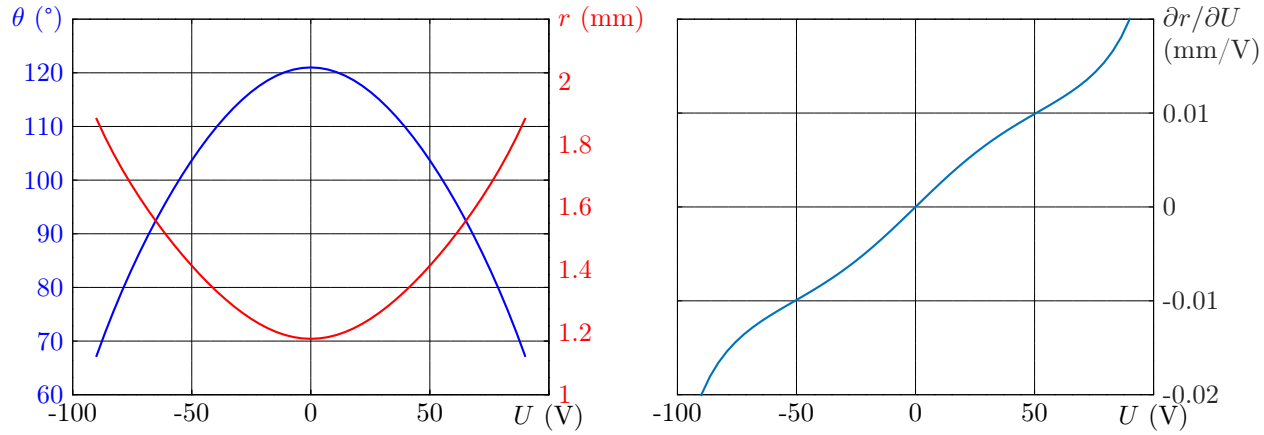

Figure S3: Calculated voltage dependence of CA θ and base radius r (left) and $\partial r/\partial U$ (right), for a small water droplet ($V = 10\,\mu\mathrm{L}$, $\gamma_{\mathrm{la}} = 72\,\mathrm{mN/m}$) placed on the Teflon AF 1600 layer ($c = 16.03\,\mu\mathrm{F/m^2}$).

In our EWOD experimental series we used a triangular voltage with a constant slope $\dot{U} = \mathrm{d}U/\mathrm{d}t = k_u$. The capillary number Ca is used to evaluate the relative influence of viscuous forces $\mu\cdot v$ (dynamic viscosity μ, characteristic velocity v) versus surface tension γ:

$\mathrm{Ca} = (\mu \cdot v)/\gamma$.

In the case of our droplet and the maximum voltage of 80 V, the maximum speed of the triple line (see Fig. S3 right) is $v_{\max} = k_u \cdot (\partial r/\partial U)_{\max} = k_u \cdot 0.02\,\mathrm{mm/V}$. With the dynamic viscosity of water $\mu \sim 0.89\,\mathrm{mNs/m^2}$ and the surface tension 72 mN/m at room temperature, $T = 25\,^{\circ}\mathrm{C}$, the capillary number is $\mathrm{Ca} = 2.47 \cdot 10^{-6} \cdot k_u$. With the maximum used voltage slope of $k_u = 16\,\mathrm{V/s}$ the capillary number $\mathrm{Ca} = 2.47 \cdot 10^{-6}$, justifies the quasi-static approach[1] evaluating the EWOD experiments.

Details of the temperature-dependent θ_{Y}

From the previous work of authors,[2] the solid surface tension can be written as:

$$\gamma_{\mathrm{s}} = \gamma_{\mathrm{s0}} + f \cdot e \quad , \tag{8}$$

where γ_{s0} is the solid surface tension in vacuum, e is the energy contribution of a fully occupied adsorption area on a solid surface, has the same unity of γ, mN/m. f is the adsorption coverage, defined as the area fraction of the occupied adsorption area to the total adsorption area. For the static, initial state of CA equilibrium ($F_{\mathrm{f}} = 0$ and $F_{\mathrm{ad}} = 0$), the solid-air interface is dry, the coverage $f = 0$, the solid-air interface tension is written as:

$$\gamma_{\mathrm{sa}} = \gamma_{\mathrm{s0}} \quad . \tag{9}$$

The solid-liquid interface is assumed as fully occupied adsorption area, namely, $f = 1$. The solid-liquid interface tension is therefore written as:

$$\gamma_{\mathrm{sl}} = \gamma_{\mathrm{s0}} + e \quad . \tag{10}$$

Plugging the Eqs. 9 and 10 into the Young's CA equation yields

$$\gamma_{\mathrm{la}} \cdot \cos\theta_{\mathrm{Y}} = -e \quad . \tag{11}$$

e is experimentally determined by our previous work, showing a constant value of 37.15 mN/m in the temperature range from 20 to 50 °C.

References

(1) Zhang, S.; Yi, Z.; Mingchao, L.; Dorian, A. H.; Yixiang, G. Dynamic contact angle hysteresis in liquid bridges. *Colloids ans Surfaces A* **2018**, *555*, 365–371.

(2) Xiang, Y.; Fulmek, P.; Platz, D.; Schmid, U. Temperature Dependence of Water Contact Angle on Teflon AF1600. *Langmuir* **2022**,