Enhancement of contact line mobility by means of infrared laser illumination

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Enhancement of contact line mobility by means of infrared laser illumination.

I. Experiments

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The shape of a droplet moving on a solid substrate is largely determined by the mobility of its contact line. Above a certain critical velocity, the droplet typically disintegrates and leaves residual liquid behind. We studied experimentally how localized infrared laser illumination can stabilize it against the progressive loss of liquid. We systematically varied the substrate velocity, the laser power, and the laser spot position and found that the critical velocity of the droplet can be substantially increased. These results are potentially important for increasing throughput in immersion lithography systems. © 2016 AIP Publishing LLC.
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I. INTRODUCTION

The motion of droplets along solid surfaces has been studied in a great variety of contexts.^{1–27} Whereas for some applications the retention of droplets is desirable, it is the removal of liquid for others. Several approaches for increasing the mobility of droplets have been investigated, such as air flow,^{28–30} mechanical vibration,^{31–33} AC electrowetting,^{34,35} lubricant films,³⁶ surface patterning,³⁷ uniform substrate heating,³⁸ and gas flow through a supporting porous substrate.³⁹

Podgorski *et al.* studied the shape of a droplet sliding down an inclined plane.¹¹ It was found that above a critical velocity, the droplet undergoes a pearling instability, where smaller droplets detach from the receding side of the droplet and are left behind on the substrate.

In this manuscript (Paper I), we introduce a non-contact technique for increasing droplet mobility that does not require any substrate modifications. We show that a localized temperature increase induced by infrared (IR) laser illumination can suppress or delay the occurrence of the pearling instability. We report on the systematic experiments characterizing the IR-induced increase of the corresponding critical velocity. In Paper II,⁵³ we present a comprehensive numerical model that elucidates the strong coupling between fluid flow, heat transfer, and contact line dynamics. In combination, the experimental and numerical results allow to determine the dominant mechanisms behind the IR-induced enhancement of the contact line mobility.

Figure 1 illustrates the experimental geometry as well as droplet shape distortions for different values of the substrate speed. A droplet is attached to a pair of stationary, vertical concentric needles, which constantly supply and extract liquid, as indicated by arrows in Fig. 1(a). The droplet is in contact with a horizontal substrate moving with velocity U_{sub} . At velocities substantially lower than the critical velocity, the receding contact line of the droplet maintains a round shape [Figs. 1(a) and 1(e)]. At velocities approaching the critical velocity U_c , the shape of the receding side of the

droplet becomes more and more acute [Figs. 1(b) and 1(f)]. At velocities above the critical velocity $U_{\text{sub}} > U_c$, the droplet loses liquid that is left behind on the substrate [Figs. 1(c) and 1(g)]. Using a laser beam of sufficient power, the receding contact line of the droplet regains a round shape and the occurrence of residual droplets is prevented.

This manuscript is organized as follows: in Section II, we describe the experimental setups. Section III presents the experimental results for variations of the substrate speed, the laser power, and the laser spot position. In Section IV, we discuss the experimental results as well as their application potential regarding immersion lithography.

II. EXPERIMENTS

A. Experimental setup—Turntable

Figure 2 shows a sketch of the experimental turntable setup. A horizontal circular polycarbonate (PC) substrate (Makrofol, Bayer, thickness 480 μm , diameter 30 cm) rests on a 3.90 mm thick glass plate, which is supported by four air-bearings (New Way, Product No. 8003941046). The plate

FIG. 1. Side-views ((a)–(d)) and bottom-views ((e)–(h)) of droplets resting on moving substrates for values of $U_{\text{sub}} \ll U_c$ ((a) and (e)), $U_{\text{sub}} \leq U_c$ ((b) and (f)), and $U_{\text{sub}} > U_c$ ((c) and (g)). The pointy morphology of the receding contact line can be smoothed and the loss of liquid prevented by IR illumination ((d) and (h)). The droplets are kept stationary at the top by means of two concentric needles. The needle stand-off height H , the receding contact angle θ_r , the opening angle ϕ , and the angle of incidence of the laser beam β are indicated.

FIG. 2. Sketch of the experimental turntable setup. The arrows labeled with “bottom view” and “side view” indicate the viewing direction of the bottom- and side view cameras. H denotes the stand-off distance of the concentric needles above the substrate surface.

is connected via a gearbox (Maxon Motor, Product No. 166182, ratio 636:1) to a motor (Maxon Motor, Product No. 136292). The angular velocity can be adjusted by a controller (Maxon Motor, Product No. hedl-5540a11). A droplet is positioned close to the perimeter of the substrate and kept in place using two concentric needles (outer diameter 2.6 mm, stand-off height $H = (0.65 \pm 0.03)$ mm). The ratio of the droplet diameter and the distance from the rotation axis is much smaller than 1. Thus, the circular motion of the substrate underneath the droplet can be considered effectively rectilinear. The droplet is monitored by means of two CCD cameras from underneath the transparent substrate and from the side, i.e., viewing radially inwards with respect to the substrate rotation.

An IR laser beam (Lumics, lu1470c020, wavelength 1470 nm, max. optical power of 20 W) coupled into an optical fiber (core diameter 400 μm , numerical aperture 0.22) illuminates the substrate and the droplet. A custom-designed optical system comprising a cylindrical lens (Thorlabs, diameter 22 mm, numerical aperture 0.138) provides an elliptical laser spot at a position close to the receding contact line of the droplet. The full width at half maximum (FWHM) along the long axis is (3.80 ± 0.02) mm, and (0.70 ± 0.01) mm along the short axis (the measurement of the laser intensity distribution is discussed in Appendix D). The angle of incidence of the laser beam is set to $\beta = 45^\circ$, such that the concentric needles do not obstruct the laser beam. The laser power was adjusted by modulating the operating signal of the laser controller with a periodic duty cycle (50 Hz), i.e., a square-wave signal with variable “on”-fraction. The operating current was maintained at a constant value of 3 A. The long axis of the ellipse is oriented in the radial direction (dotted line in Fig. 2) with respect to the substrate rotation axis. The position of the laser spot on the substrate was calibrated using a thermocouple (Omega, CHAL-0005, head diameter 13 μm).

The working liquid is ethylene glycol (EG), which is partially wetting on PC. EG has a relatively low vapor pressure in order to minimize evaporation effects. Moreover, the critical velocity of ethylene glycol is in the accessible range of the turntable setup. Temperature dependent material parameters of EG are discussed in Appendix A. The surface

roughness of the PC substrates was characterized by atomic force microscopy (AFM) as described in Appendix B. Additional details of the turntable setup, such as the layout of the concentric needles, the optical system, and the laser intensity distribution, as well as the data analysis procedures, are also described in the Appendices.

B. Experimental setup—Contact angle (CA) measurements

The quasi-static advancing and receding CAs were determined by slowly increasing or decreasing the volume of a sessile droplet until the contact line starts to advance or recede, respectively. For these measurements, a PC substrate is placed on a hotplate, and a droplet of EG is deposited on it using a syringe needle, which remains submerged in the liquid. The needle is connected to a syringe pump (KDS 88) that slowly adds or removes liquid to/from the droplet. The droplet is imaged from the side with a CCD camera (Thorlabs, dcc1645c) and illuminated from the opposite side. An aluminium enclosure with two glass windows is placed over the substrate and the droplet to minimize evaporation effects and to improve and accelerate thermal equilibration. The contact line speed was (26 ± 4) $\mu\text{m/s}$ during the measurements.

III. EXPERIMENTAL RESULTS

A. Quasi-static contact angles

We measured the temperature-dependence of the CAs of EG on PC using a custom-built thermostatted CA-goniometer. Figure 3 shows the experimental results for the quasi-static advancing (circles) and receding CAs (squares). The temperatures reported are the set values on the hotplate controller. The solid lines are guides to the eye. The advancing and receding CAs slightly increase and decrease with temperature, respectively. Petke and Ray found that the quasi-static advancing and receding CAs vary approximately linearly with temperature⁴⁰ with rates of -0.03 deg/K and 0.12 deg/K, respectively. The straight lines in Fig. 3 have slopes of (0.04 ± 0.02) deg/K and (-0.05 ± 0.03) deg/K, respectively, i.e., our values have similar magnitude but opposite signs compared with Petke and Ray’s results. One possible reason could be different surface conditions of the PC substrates.

FIG. 3. The quasi-static receding and advancing contact angles as a function of temperature measured on a stationary substrate.

B. Variation of laser power

First, we studied the influence of the laser power P on the shape of the droplet which was parameterized by the receding CA θ_r and the opening angle φ of the receding contact line, as depicted in Figs. 1(b) and 1(f). The substrate velocity was kept constant at $U_{\text{sub}} = (4.28 \pm 0.05)$ mm/s, which corresponds to a capillary number of $\text{Ca} \equiv \mu U_{\text{sub}} / \gamma = 1.6 \times 10^{-3}$, using values of μ and γ at $T = 24^\circ\text{C}$. The distance d between the outer edge of the needle on the receding side and the center of the laser spot was kept constant at 0.50 mm [see Fig. 4(c)]. Figures 4(a)–4(c) show bottom-views of a droplet for different laser powers. The red-shaded areas in Fig. 4(c) indicate the laser intensity levels of $\geq 43\%$ and $\geq 72\%$ of the maximum value. The droplet shape is pointed at the receding side when the laser is off due to the high substrate velocity [Fig. 4(a)] and obtains a progressively more circular shape for increasing laser powers [Figs. 4(b) and 4(c)]. Figures 4(d)–4(f) show side-views of a droplet for

FIG. 4. (a)–(c) Bottom-views and (d)–(f) side-views of an EG droplet for $U_{\text{sub}} = (4.28 \pm 0.05)$ mm/s, $d = 0.5$ mm, and different laser powers P . (g) and (h) The influence of the laser power on (g) the opening angle φ and (h) the receding contact angle θ_r for constant values of $U_{\text{sub}} = (4.28 \pm 0.05)$ mm/s and $d = 0.50$ mm. The continuous lines are guides to the eye.

different laser powers. For clarity, the needle is overlaid with a white square. The droplet is pinned at the outer edge of the outer needle. The dotted line indicates the location of the substrate. Below this line, the reflection of the droplet in the substrate is visible.

Figures 4(g) and 4(h) present φ and θ_r as a function of P , which both increase with laser power. A larger value of φ reflects a rounder, more circular droplet shape. For $P > 2.5$ W, the opening angle φ approaches a constant value, while θ_r increases over the entire power range. The vertical error bars are primarily caused by weak surface heterogeneities, contamination, and defects that induce variations of the receding contact angle and the contact line shape.

C. Variation of the laser spot distance

Next, we studied the influence of the laser spot distance d . Figures 5(a)–5(c) show bottom-view images of EG droplets for $P = (1.87 \pm 0.02)$ W and $U_{\text{sub}} = (4.28 \pm 0.05)$

FIG. 5. (a)–(c) Bottom-views and (d)–(f) side-views of EG droplets for $P = (1.87 \pm 0.02)$ W, $U_{\text{sub}} = (4.28 \pm 0.05)$ mm/s, and different laser spot distances d . (g) and (h) Opening angle φ (g) and receding contact angle θ_r (h) as a function of laser spot distance d for $P = (1.87 \pm 0.02)$ W and $U_{\text{sub}} = (4.28 \pm 0.05)$ mm/s. The solid lines are guides to the eye.

mm/s and different values of d . The droplet shape is pointed when the laser is either turned off or positioned at a large distance from the receding contact line. For small values of d , the droplet shape becomes rounder and the droplet length decreases. This decrease of droplet length can also be observed in the side-view images in Figs. 5(d)–5(f). Figures 5(g) and 5(h) show φ and θ_r as a function of d , including their values when the laser is off. For increasing d , both parameters approach their off-value. The rate of change diminishes for $d \geq 1.4$ mm, which equals approximately 1.5 times the FWHM of the laser spot $\beta = 45^\circ$, i.e., for IR-laser illumination to be effective, the laser beam must be positioned sufficiently close to the contact line.

D. Variation of substrate speed

Finally, the combined influence of U_{sub} and P on the droplet shape is presented in Fig. 6. Figures 6(a) and 6(b) show bottom-views of unirradiated droplets (i.e., $P = 0$ W) for $U_{\text{sub}} \approx 2$ mm/s and 5 mm/s. After switching on the IR laser at different power settings, the substrate velocity has been progressively increased, while monitoring the receding contact angle θ_r and the opening angle φ , as shown in Figs. 6(c) and 6(d). The procedure was stopped as soon as residual liquid droplets were left behind on the substrate. The last data points in Figs. 6(c) and 6(d), i.e., those with the highest value of U_{sub} in each series, are the highest substrate speeds that did not induce droplet shedding. The vertical arrows in Fig. 6(c) indicate the approximate values of the corresponding critical velocities U_c .

FIG. 6. (a) and (b) Droplet bottom-views for $P = 0$ W and a substrate speed of (a) $U_{\text{sub}} \approx 2$ mm/s and (b) 5 mm/s. (c) and (d) Droplet opening angle φ and receding contact angle θ_r as a function of U_{sub} for $d = (0.50 \pm 0.01)$ mm and four different values of the laser power $P = 0, 0.6, 1.9$, and 3.7 W.

The opening angle is constant for small substrate velocities and decreases for higher substrate velocities. A smaller opening angle corresponds to a more acute contact line shape. The substrate velocity at which the opening angle starts to decrease increases with laser power. Residual droplets are left behind on the substrate once φ is smaller than approximately 60° . Figure 6(d) shows the receding contact angle θ_r as a function of U_{sub} . The receding contact angle continuously decreases for an increasing substrate velocity to a terminal value of approximately $(5 \pm 3)^\circ$.

Figure 7 shows the relative increase of the critical velocity U_c as a function of laser power P . The critical velocity increases by a factor of almost 2 when applying a laser power of $P \approx 4$ W.

IV. DISCUSSION

The influence of non-uniform temperature distributions on a receding contact line could be due to the following three phenomena. First, temperature gradients induce gradients in the surface tension, which gives rise to thermocapillary stresses.^{41,42} These stresses can provide an additional driving force for contact line motion. Second, an increased temperature results in the reduction of the liquid's viscosity, i.e., a reduction in viscous friction. Finally, the temperature dependence of the liquid-solid, the liquid-vapor, and the solid-vapor surface tensions may cause changes in the contact angle, as expressed by Young's law.^{40,43}

In order to evaluate the third option, we performed the measurements of the temperature-dependent CAs as shown in Fig. 3. Considering the measurement uncertainty of the CA data and the small numerical values of the rates, we conclude that the CAs are essentially independent of temperature and consequently do not affect the droplet mobility strongly. Moreover, Fig. 6(d) shows that θ_r increases with laser power and for decreasing values of U_{sub} . Both effects tend to increase the temperature rise and according to Fig. 3, a smaller value of θ_r would be expected, contrary to the experimental results.

Winkels⁴⁴ measured the shape of *isothermal* sliding droplets as a function of speed and found that the opening angle at the critical substrate speed equalled $\varphi = 60^\circ$. To good approximation, the same value of $\varphi \approx 60^\circ$ is also observed in Fig. 6(c) at the termination points of the curves,

FIG. 7. The relative increase of the critical substrate velocity as function of laser power. The solid line is a guide to the eye.

FIG. 8. The temperature-dependency of the viscosity (solid black line), surface tension (dotted red line), and mass density (dashed blue line) of ethylene glycol, normalized by their corresponding values at 20 °C.

i.e., at the respective critical speeds. This is an indication that Marangoni stresses are probably not the dominant mechanism behind the substantial increase of the critical speed, but rather the thermally induced decrease in viscosity. Figure 8 illustrates the strong temperature dependence of μ .

In Fig. 6(d), it is observed that θ_r at low speeds $U_{\text{sub}} \approx 0.2 \text{ mm/s}$ increases with increasing laser power and thus with increasing temperature. Using a very similar setup, Riepen measured the dynamic receding contact angle of a droplet stabilized by continuous in- and outflow through concentric needles as a function of substrate speed.^{28,29} There is one crucial difference, however, compared with our experiments: in Riepen's experiments, the droplet contact line was pinned at the *inner* needle, whereas in our experiments it was pinned at the *outer* needle. Consequently, the extraction flow through the outer needle caused a significant air flow near the receding contact line, which in turn induced an inwards-oriented shear stress at the liquid-air interface. Riepen observed that the receding contact angle was systematically higher *with* airflow than without airflow. This is consistent with our interpretation that inwards-oriented Marangoni flow tends to increase θ_r . However, in our case, the magnitude of the shear stress is coupled to the substrate speed and diminishes with increasing U_{sub} . In the experiments by Riepen, the shear stress was approximately independent of U_{sub} , and thus the effect was observed for the entire range of substrate speeds.

A comprehensive and conclusive investigation into the dominant mechanism behind the contact line mobility enhancement is presented in Paper II.

A. Application perspectives

The achievable resolution in immersion lithography can be improved by introducing a liquid in between the objective lens and the photoresist-covered semiconductor wafer.^{45,46} The overall throughput benefits from a high velocity of the wafer relative to the projection optics. However, above a critical velocity liquid is entrained on the surface, which negatively affects the lithography process.^{28,29} This entrainment-induced occurrence of residual liquid could be

suppressed by IR irradiation. Compared with the IR power levels reported in this manuscript, the required laser intensity can be substantially reduced by more tightly focusing the laser at the apex position of the receding contact line and by selection of a laser wavelength close to a maximum of the optical absorption of the liquid.

The enhancement of the contact line mobility is also the key mechanism that allows so-called “clean” dewetting of thin liquid films, i.e., without occurrence of residual droplets, by means of IR-laser irradiation with potential applications in large-area electronics and solution processing of organic electronic devices.⁴⁷

V. SUMMARY

In this manuscript, we studied a non-contact technique for increasing the mobility of stationary droplets on moving, partially wetting substrates that does not require any substrate modification. We developed an experimental setup where a droplet, which is attached to concentric needles that constantly supply and extract liquid, is positioned on a rotating turntable. An elliptical infrared laser spot is used to locally increase the temperature in the vicinity of the receding contact line. The deformation of the droplet is monitored by two cameras in side- and bottom-view perspectives. We determine the receding contact angle and the shape of the contact line as a function of substrate speed and laser illumination.

Above a certain critical velocity, the droplet typically disintegrates and leaves residual liquid behind. We found that by means of infrared laser illumination, the critical velocity of the droplet can be substantially increased and the occurrence of residual liquid left behind on the substrate can be suppressed. *A priori*, it was unclear whether the observed effect was caused by a thermocapillary shear, a temperature-induced reduction of viscosity, or a change of the equilibrium contact angles. The third option was eliminated by measurements of the quasistatic advancing and receding contact angles, which exhibited only a minute temperature dependence for our material system. In Paper II, we present a comprehensive numerical model that elucidates the strong coupling between fluid flow, heat transfer, and contact line dynamics. In combination, the experimental and numerical results allow us to determine the dominant mechanisms behind the IR-induced enhancement of the contact line mobility.

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APPENDIX A: MATERIAL PROPERTIES

The viscosity μ and surface tension γ of EG can be expressed^{48,49} as functions of temperature T [in degrees Celsius] as follows:

$$\mu [\text{mPa s}] = \exp(-3.61 + 986.52/(T + 127.86)), \quad (\text{A1})$$

$$\gamma [\text{mN/m}] = 50.21 - 0.09 T, \quad (\text{A2})$$

which are plotted in Fig. 8, normalized by their values at $T = 20^\circ\text{C}$. The viscosity of EG shows by far the largest variation as function of temperature. The latter has also been measured as $(17.6 \pm 0.3) \text{ mPa s}$ using a Brookfield LVDV-II+PRO viscometer at a temperature of 24°C , which compares favorably with the value 17.86 mPa s calculated from Eq. (A1).

The thermal conductivity of EG is $k = 0.24 \text{ W/(m K)}$ and its heat capacity $c_p = 2.3 \text{ kJ/(kg K)}$,⁴⁹ resulting in a thermal diffusivity of $\kappa = k/(\rho c_p) = 1.08 \times 10^{-7} \text{ m}^2/\text{s}$. The thermal conductivity of PC is $k_{\text{sub}} = 0.22 \text{ W/(m K)}$, the mass density $\rho_{\text{sub}} = 1200 \text{ kg/m}^3$, and its heat capacity $c_{p,\text{sub}} = 1240 \text{ J/(kg K)}$, resulting in a thermal diffusivity of $\kappa_{\text{sub}} = 1.48 \times 10^{-7} \text{ m}^2/\text{s}$. The material parameters of glass⁵⁰ were assumed to be $\rho_{\text{glass}} = 2550 \text{ kg/m}^3$, $c_{p,\text{glass}} = 750 \text{ J/(kg K)}$, and $k_{\text{glass}} = 1 \text{ W/(m K)}$, resulting in a thermal diffusivity of $\kappa_{\text{glass}} = 5.2 \times 10^{-7} \text{ m}^2/\text{s}$.

APPENDIX B: AFM MEASUREMENTS OF SURFACE ROUGHNESS

The surface roughness of the PC substrates was analyzed using AFM (Bioscope Catalyst fitted with DNP-10 tips). The substrates are supplied with protective foils on both sides, which are adhering by means of electrostatic charge. The protective foil was removed from all samples and the polycarbonate samples were decharged using an anti-static bar (Simco-Ion MEB) before the measurement. The AFM data presented in Figs. 9(a) and 9(b) are flattened and tilt-compensated. The height profiles along the dashed lines are represented in the line graphs right of the images. An AFM image of a PC substrate (Sabic Lexan 9030, not used

FIG. 9. Atomic force microscopy images and cross-sections for (a) PC, Sabic Lexan 9030 and (b) PC, Bayer Makrofol DE1-1CC. (Data taken from Ref. 51.)

in this study) is presented in Fig. 9(a). The AFM scan shows horizontal trenches with a depth of up to 10 nm and a spacing of approximately $8\text{--}9 \mu\text{m}$. Perpendicular to these trenches, there are irregularly spaced vertical ridges with a height of approximately 30 nm. An AFM image of another PC substrate (Bayer Makrofol DE1-1CC, used in this study) is shown in Fig. 9(b). Some randomly oriented ridges are visible on the surface, which exhibits more gradual fluctuations with a length scale of $20\text{--}40 \mu\text{m}$ and an amplitude of approximately $\pm 8 \text{ nm}$.

APPENDIX C: SUPPLY AND EXTRACTION OF LIQUID

The concentric needles consist of two hollow concentric cylinders with separate fluid connections. Figure 10 shows a bottom-view microscope image of the concentric needle. The different radii of the two cylinders are indicated. The small cylinder has an inner radius of $r_1 = (0.41 \pm 0.01) \text{ mm}$ and an outer radius of $r_2 = (0.60 \pm 0.01) \text{ mm}$. The large cylinder has an inner radius of $r_3 = (0.79 \pm 0.01) \text{ mm}$ and an outer radius of $r_4 = (1.28 \pm 0.01) \text{ mm}$.

Liquid is supplied at constant flow rate through the inner cylinder, while the outer cylinder constantly extracts a liquid-air mixture. The liquid in the droplet is therefore continuously refreshed. The arrows superimposed on the needle in Fig. 1(a) indicate the flow directions. The liquid supply is contained in a jar positioned approximately 40 cm above the setup. Gravity provides the supply pressure and the flow rate is controlled using a valve. The extracted mixture is pumped into a gas-liquid separator by means of a vacuum pump (KNF, Product No. N811KT.18) connected to the air outlet of the separator. A pressure regulator (SMC, Product No. irv2000-F02BG) controls the pressure in the separator. A droplet is obtained and stabilized by iteratively adjusting the supply and extraction rates. Typically, the supply rate is set to $(7 \pm 1) \text{ ml/min}$. The pressure difference at the separator is set to $(-8.0 \pm 0.5) \text{ kPa s}$, resulting in an air flow of $(1.5 \pm 0.5) \text{ l/min}$.

APPENDIX D: LASER BEAM PROFILE MEASUREMENTS

The intensity profile I of the infrared laser beam has been measured using a scanning slit setup. A metal plate with a small slit (width $(55.8 \pm 0.4) \mu\text{m}$) has been placed

FIG. 10. Microscope bottom-view image of the concentric needles. The inner and outer radii of the inner and outer hollow cylinder are denoted by r_1 and r_2 as well as r_3 and r_4 , respectively.

above a power sensor (Thorlabs, s314c), which was translated at a constant speed of $20 \mu\text{m/s}$ by means of a motorized linear translation stage (Standa, 8MTF). This low speed was chosen owing to the relatively slow response of the sensor.

Figure 11 shows the measured beam profiles for $\beta = 0^\circ$ and 45° along the long and short axes, respectively. The solid lines correspond to fit-functions of the form

$$I = c_0 \exp\left(\frac{-2x'^2}{c_1}\right), \quad (\text{D1})$$

with fit parameters c_0 and c_1 , which are excellent approximations to the experimental data. The FWHM along the short axis for an angle of incidence $\beta = 45^\circ$ was 0.96 mm , which is much larger than the slit width.

APPENDIX E: FLUCTUATIONS OF THE STAND-OFF DISTANCE

A fresh substrate has been used for every series of experiments. The substrates are supplied with protective foils on both sides, which are adhering by means of electrostatic charge. First, the protective foil on the bottom side of the substrate is removed and the substrate is placed on the glass plate. Due to the small aspect ratio (PC substrate thickness $480 \mu\text{m}$, substrate diameter 30 cm), however, air bubbles were frequently trapped underneath the substrate. In order to squeeze out air trapped between the plates, we used a soft roller. Subsequently, the protective foil on the top side is removed. Static electricity from both the substrate and the glass plate is removed using a static control bar (Simco-Ion, MEB). The glass plate is subsequently placed on the air-bearings and connected to the motor.

An important mechanical characteristic of turntables is the stability of the stand-off distance H . We used a confocal sensor (MICRO-EPSILON, IFS 2405) to measure variations $H \pm \Delta H$ of the vertical position of the moving substrate with respect to the (non-moving) concentric needles at the position of the needles. This sensor measures the absolute distance from the sensor to the top surface of the substrate, while the substrate is rotating. Typical variation amplitudes ΔH were found to be $50 \mu\text{m}$ when the PC substrate was

initially placed on the supporting glass plate. After removal of the air bubbles by means of a soft roller, ΔH was typically $10\text{--}15 \mu\text{m}$, which is much smaller than $H \approx 500 \mu\text{m}$.

APPENDIX F: DETERMINATION OF THE DROPLET SHAPE

Two cameras record the droplet shape simultaneously from the bottom (Allied Vision Technologies, Pike F-145B) and from the side (Allied Vision Technologies, Guppy F-146B). The bottom of the droplet is imaged through the transparent glass plate and substrate and is illuminated along the camera's viewing direction. The viewing direction of the side-view camera is radially inwards with respect to the circular substrate. Both cameras are protected against the infrared radiation with a short-pass filter (Schott KG-3), which is transparent for visible light. The frame rate of both cameras is set to 7.5 frames per second. The magnifications of the optics used for the bottom- and side-view images were $(4.91 \pm 0.02) \mu\text{m/px}$ and $(9.22 \pm 0.04) \mu\text{m/px}$, respectively.

From the side-view images, we extracted the receding contact angle θ_r . From the bottom-view images, we extracted the radius of curvature R and the opening angle φ of the tip. In both cases, we used edge detection routines in MatLab based on a Canny filter,⁵² to determine the (x, y) pixel-pairs corresponding to the perimeter of the droplet.

The bottom-view images are first cropped and transformed into black and white. The curvature radius R is determined by fitting a circle through the apex of the droplet contact line. Figures 12(a) and 12(c) show the pixels of the droplet contact line for 2 representative cases of a round and a pointed contact line shape. The pixel with the largest y -coordinate value is identified as the apex point. All pixels within 10 pixels of the apex in the negative y -direction [indicated by blue squares in Figs. 12(a) and 12(c)] are used to fit a circle, the radius of which is denoted R . Figures 12(b) and 12(d) show the fitted curvature circles.

The opening angle is determined by fitting of two straight tangent lines through the side of the droplet. A range of 10 pixels in the x -direction above or below the first points

FIG. 11. The measured integrated intensity profiles of the IR laser beam for angles of incidence $\beta = 0^\circ$ and 45° along the long and short axes, respectively. The solid lines correspond to fit-functions according to Eq. (D1).

FIG. 12. Contact line shape at the tip of the droplet tail extracted from typical bottom-view images for (a) a round and (c) a pointed droplet shape, corresponding to substrate speeds $U_{\text{sub}} \ll U_c$ and $U_{\text{sub}} \approx U_c$, respectively. The fitted circles (red dotted line) and tangent lines (black solid lines) are, respectively, shown in (b) and (d).

not used for the circle fit [indicated by red squares in Figs. 12(a) and 12(c)] is used to fit the straight lines. The opening angle φ of the contact line is determined from the slopes of the two lines. Figures 12(b) and 12(d) show the fitted tangents. Due to the restriction to a range of 10 pixels (beyond the pixels used for fitting the curvature radius), it is evident that (1) even for a perfectly circular contact line of radius R , the extracted value of φ will remain below 180° and (2) the exact value of φ (approx. 155° in Fig. 6 at low U_{sub}) depends on R (and thus the droplet volume and outer needle diameter). This is, however, not an impediment as the opening angle is a sensible parameter only for pointed contact line shapes corresponding to larger values of U_{sub} .

The receding contact angle is determined similarly by fitting a straight line through the tip of the droplet extracted from side-view images. The pixel with highest y -coordinate is identified as the apex of the droplet. All points within 10 pixels of the tip along the interface are used to fit a straight line. The receding contact angle is calculated from the slope of this line.

The data for θ_r and φ in Figs. 4, 5, and 6 represent averages determined from 100 camera frames. The error bars correspond to the standard deviation of these sets of 100 datapoints.

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