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Citation for published version (APA):

DOI:
10.1016/j.jcis.2013.06.011

Document status and date:
Published: 01/01/2013

Document Version:
Publisher’s PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

• A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
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Deformation and dewetting of thin liquid films induced by moving gas jets

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ARTICLE INFO

Article history:
Received 23 April 2013
Accepted 6 June 2013
Available online 26 June 2013

Keywords:
Thin liquid films
Dewetting
Film rupture
Thin film instability
Impinging jets

ABSTRACT

We study the deformation of thin liquid films subjected to impinging air-jets that are moving with respect to the substrate. The height profile and shape of the deformed liquid film is evaluated experimentally and numerically for different jet Reynolds numbers and translation speeds, for different liquids and substrate materials. Experiments and numerical results are in good agreement. On partially wetting substrates film rupture occurs. We imaged the appearance of dry spots and emergence of droplet patterns by high-speed, dual-wavelength interference microscopy. We systematically evaluated the resulting average droplet size and droplet density as a function of the experimental conditions. We show that within experimental accuracy the distribution of dry spots is dependent only on the residual film thickness and is not directly influenced by the shear stress and pressure gradients of the air-jet, nor by the speed of the substrate.

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1. Introduction

The interaction between gas jets and moving liquid films is an important aspect of many technological processes. In the galvanization of steel, for example, so-called jet stripping is a common technique [1–8], where the thickness and uniformity of a metallic coating on a moving sheet of steel is adjusted by the pressure and shear stress of an impinging slot jet. The analogous effect is also relevant in smaller scale systems, such as ultrathin films of hard disk lubricant [9]. Moreover, gas jets were employed to study the influence of surfactants on the propagation of surface waves in a liquid film on a rotating disk [10].

When an impinging jet deforms a liquid film on a partially wetting substrate, the film can rupture [11–13] and start to dewet. Consequently air-jets are employed to initiate the redistribution of liquid in continuous coating of chemically patterned surfaces [14]. In immersion lithography [15–17], a photoresist covered silicon wafer is exposed through a layer of water in order to increase the effective numerical aperture of the illumination system. Controlled air-flows are utilized to contain the water meniscus between the objective lens and a partially wetting wafer, which is moving at a relative speed of approximately 1 m/s. Above a critical translation speed, however, a thin liquid film is left on the wafer, that spontaneously breaks up, dewets and leads to undesirable residual droplets on the substrate. Similar film rupture and dewetting phenomena have been described for thin layers of polymer melts [18–27], metals [28–32] and oil films submersed in water [33–35].

In this paper, we present a systematic study of the deformation of a moving liquid film due to laminar air-jets emanating from a round nozzle. We studied the behavior of four different liquids on both wetting and partially wetting substrates. We performed quantitative experiments and numerical simulations elucidating the dependence of the shape and minimum thickness of the deformed film on the operating parameters. The results were rationalized by means of a scaling analysis in the limit of high jet Reynolds numbers. On partially wetting substrates, we identified the minimum film thickness to be the crucial parameter governing the breakup process. We systematically varied the substrate speed and jet Reynolds number and determined the densities of dry-spots and residual droplets as well as the droplet size distribution. Using a geometrical scaling analysis [20], we derived power law relations for the droplet density and average droplet radius. Depending on the ratio of the dominant instability wavelength and the transverse lengthscale of the film deformation, a transition from two-dimensional to quasi-one-dimensional rupture patterns was observed.

2. Experiments

Fig. 1 shows a schematic representation of the experimental setup. A thin liquid film of initial thickness \( h_0 \) on a transparent substrate is translated with respect to an air-jet that impinges at normal incidence. The air-jet is generated by maintaining an air flow \( Q_{air} \) through a round, hollow needle of inner diameter \( D \). Its orifice is positioned at a distance \( H \) above the thin liquid film. A local depression, which we call track, is formed in the liquid film along the jet trajectory as shown in Fig. 2a. The liquid displaced from
For these liquids and their aqueous solutions, the apex of the front rim is called the ‘track’. The maximum lateral extension of the rim is termed the track width \( w_{\text{max}} \). The distance between the jet stagnation point and the centerline of the jet trajectory is accumulated in a rim ahead of the track. The maximum lateral extension of the rim is termed the track width \( w_{\text{max}} \). The distance between the jet stagnation point and the centerline of the jet trajectory is accumulated in a rim ahead of the track. The maximum lateral extension of the rim is termed the track width \( w_{\text{max}} \).

For the preparation of the thin liquid film and translation of the sample with respect to the impinging jet, we used a custom-designed spin-coater, where the sample and the area around the axis of rotation is optically accessible from both sides. Our spin-unit consists of a rotating sample holder that is supported by a hollow cylindrical roller bearing and driven by a tooth belt and a precision motor. Rotation speeds range between 0 and 6000 rpm with a resolution of 0.2 rpm. Transparent substrates were attached to the rotation stage using double-sided adhesive tape at the corners (3M Scotch 4016-1/2). As wetting substrates we used microscope cover glasses (Gold Seal, 48 mm, Cat#63774-01), cleaned and rendered fully wettable by UV ozone treatment (Jelight) for 20 min. Partially wetting substrates were optical-quality polycarbonate film (PC, thickness 750 µm, cut to the dimensions of 60 × 60 mm), which we used as received after removal of the electrostatically bound protection foil. The non-zero conductivity of the liquid ensured that any residual static electricity was dissipated when the liquid was dispensed prior to spin coating.

Liquids used in this study were glycerol (Fluka, product number 49770, purity ≥99.5%), triethylene glycol (3EG, Sigma, product number 95126, purity ≥99%), ethylene glycol (1EG, J.T. Baker, product number 8201, MOS grade) and deionized water (Smart2-Pure, TKA, resistivity 18.2 MΩ cm). Table 1 presents viscosity \( \nu \), surface tension \( \gamma \), the typical refractive index for visible wavelengths \( n \) and the dielectric constant \( e_\varepsilon \) for these liquids and their contact angles on polycarbonate. Advancing and receding contact angles on PC were measured by slowly increasing and decreasing the volume of a liquid droplet and capturing sideview images using a CMOS camera (Thorlabs DC1C645C) and telecentric lens (Moritex MML2-ST65). Unless specified otherwise, the experimental initial film thickness has the value listed in Table 1.

After spin coating, the rotating sample stage was decelerated to the rotation speed required for the impinging jet experiment. The air-jet was generated using a stainless steel hollow needle with inner diameter \( D = 208 \) µm and turned on by switching a two-way solenoid valve after reaching the required sample rotation speed. The needle was positioned at a distance \( H \) above the air–liquid interface at a radial distance of approximately 1 cm from the rotation center of the sample stage. The air (Linde, Nz/O2-mixture, humidity <200 vpm, oil-free) was supplied from a gas cylinder through a precision pressure regulator (Norgren 11-818) and filters (Headline, 25-64-50C, 99.99+% removal of 0.1 µm particles and aerosols) before being admitted to a needle valve (Brooks NRS8514) that controlled the flow rate. The mass flow was measured using a mass flow meter (Bronkhorst F-201CV-2K).

The time evolution of the height profile of the liquid film was measured through the substrate by interference microscopy using two different wavelengths. The light source consists of two high-power light-emitting diodes (LED, Luxeon I) with center wavelengths of 466 nm and 655 nm and full width half maximum (FWHM) values of the light output spectrum of 28 nm and 22 nm, respectively. The LEDs were powered by two DC power supplies (Delta Electronics, ES030-5), collimated and combined using a 50:50 beamsplitter. The light sources were modulated in synchronization to the framerate of the camera, such that the illumination wavelength was alternating from frame to frame. For low-speed imaging at a framerate of 17 frames per second (fps), we used an InfiniTube lens system with a microscope objective (Mitutoyo, MPlan APO 2×/NA = 0.055) and a CCD camera (Guppy, Allied Vision Technologies) and synchronized the light sources using Labview software and a data acquisition unit (National Instruments USB-6008). For imaging at 250 fps, we used a high-speed camera (Photron SA-4) fitted with a lens system and a microscope objective (Olympus, UPlan 4×/NA = 0.13) in combination with a dual-channel function generator (Yokagawa FG120) to synchronize the light sources. The deformed height profile of the liquid film was determined from the position and number of the interference fringes, whereby the fringe order was established by a comparison of the interference patterns of both wavelengths. The minimum film thickness near the center of the jet impingement zone was estimated by grayscale interpolation for both wavelengths. For the material systems used, the accuracy of this method is typically better than 10 nm.

Fig. 2 illustrates the dual-wavelength interference technique for a typical experiment. In Fig. 2 the blue1 and red interference micrographs are overlayed. In this particular example, a dust fiber on the

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1 For interpretation of color in Figs. 2 and 11, the reader is referred to the web version of this article.
back of the substrate is visible in the left upper corner, revealing the substrate displacement between the two consecutive frames (17 fps, \( \Delta t = 60 \) ms). The thin film deformation had already reached a steady state. The film thickness profile in Fig. 2b is based on the cross-section indicated by the dashed line in Fig. 2a. Blue and red symbols indicate the position of dark and bright fringes of the two wavelengths, whereas the symbols at \( x = 0 \) represent the minimum film thickness \( h_{\text{min}} \) obtained by grayscale interpolation for each respective wavelength. To record the droplet patterns resulting from experiments on partially wetting substrates with optimal contrast, we imaged the samples with an Olympus BX51 microscope using different magnifications between 2.5 \( \times \) and 20 \( \times \) and a CCD camera (Pike, Allied Vision Technologies).

3. Numerical models

We modeled the deformation of a thin liquid film on a substrate, moving with respect to a stationary impinging jet. Our model combines the shear stress and pressure distributions of the impinging jet with a thin film model based on the lubrication approximation. The movement of the substrate is approximated by a linear motion with a velocity \( U_{\text{sub}} \) in the \( y \)-direction, which is valid in the case that the impingement zone is sufficiently far away from the axis of rotation. The two-dimensional computational domain is sketched in Fig. 3a along with the applicable boundary conditions. To obtain the pressure and shear stress distributions in Cartesian coordinates, we converted the results of the axisymmetric simulations according to

\[
P_{\text{jet}}(x, y) = P\left(\sqrt{x^2 + y^2}\right),
\]

\[
\tau_x = \tau_{\text{jet}} \left(\sqrt{x^2 + y^2}\right) \cos \phi,
\]

\[
\tau_y = \tau_{\text{jet}} \left(\sqrt{x^2 + y^2}\right) \sin \phi,
\]

where \( \phi = \arctan(y/x) \).

We used symmetry boundary conditions at \( x = 0 \) and \( x = L_x \)

\[
\frac{\partial P}{\partial x} = 0 \quad \text{and} \quad \frac{\partial h}{\partial x} = 0,
\]

and prescribed the flux \( Q_y \) at the boundaries in the \( y \)-direction to account for the moving substrate. At \( y = -a L_y \), an undisturbed film of thickness \( h_0 \) is entering the domain

\[
Q_y = U_{\text{sub}} h_0 \quad \text{at} \quad y = -a L_y,
\]

while liquid is exiting the domain at \( y = L_y \)

\[
Q_y = U_{\text{sub}} h \quad \text{at} \quad y = L_y.
\]

Table 1

<table>
<thead>
<tr>
<th>Liquid</th>
<th>( \mu ) (mPa s)</th>
<th>( \gamma ) (mN m)</th>
<th>( \theta_{\text{adv}} ) (°)</th>
<th>( \theta_{\text{rec}} ) (°)</th>
<th>( n ) (–)</th>
<th>( \alpha ) (–)</th>
<th>( P_{\text{max}} ) (Pa)</th>
<th>( h_{\text{b}} ) (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glycerol</td>
<td>1000</td>
<td>63.0</td>
<td>81 ± 5</td>
<td>54 ± 3</td>
<td>1.48</td>
<td>42.5</td>
<td>&lt;0.1</td>
<td>17</td>
</tr>
<tr>
<td>Triethylene glycol (3EG)</td>
<td>49</td>
<td>45.5</td>
<td>67 ± 2</td>
<td>30 ± 2</td>
<td>1.46</td>
<td>23.7</td>
<td>&lt;1</td>
<td>5.0</td>
</tr>
<tr>
<td>Ethylene glycol (1EG)</td>
<td>16</td>
<td>48.0</td>
<td>66 ± 2</td>
<td>33 ± 5</td>
<td>1.43</td>
<td>37.0</td>
<td>7.7</td>
<td>4.7</td>
</tr>
<tr>
<td>Water (H₂O)</td>
<td>1</td>
<td>72.0</td>
<td>99 ± 2</td>
<td>59 ± 2</td>
<td>1.33</td>
<td>80.1</td>
<td>2300</td>
<td>6.0</td>
</tr>
</tbody>
</table>

Fig. 3a summarizes the boundary conditions and shows the mesh of the two-dimensional domain. We typically used a domain of \( 4 \times 4 \) mm², i.e., \( L_x = 4 \) mm, \( L_y = 3 \) mm and \( a = 1/3 \) or 2/3. The origin
coincides with the stagnation point of the impinging air-jet. We used the finite element software COMSOL 3.5a to solve Eqs. (1)–(4) using the initial condition $h = h_0$ until a steady-state profile was reached.

### 4. Results and discussion

Fig. 3b shows an example of a steady state height profile corresponding to $Re_0 = 500$ and $U_{sub} = 2$ mm/s. Here, the jet Reynolds number $Re_0$ is defined as $Re_0 = \frac{\rho_{gas} v D}{\eta_{gas}}$. In the following sections, we will systematically investigate how the minimum film thickness $h_{min}$ and the trend width $w_{min}$ depend on jet Reynolds number $Re_0$ and substrate speed $U_{sub}$. Moreover, we study the characteristics of dry-spot formation and residual droplet patterns as a function of $Re_0$ and substrate speed $U_{sub}$.

#### 4.1. Minimum film thickness $h_{min}$

We measured and simulated the steady state deformation of different liquids as a function of $Re_0$ and $U_{sub}$. In Fig. 4, we show the dependence of the minimum film thickness $h_{min}$ on these parameters. Fig. 4a shows the influence of $Re_0$ for 3EG films for different values of $U_{sub}$. In Fig. 4b, similar graphs are shown for glycerol, 1EG and water. Fig. 4c shows the dependence of $h_{min}$ on $U_{sub}$, whereas Fig. 4d contains all data from Fig. 4a–c in non-dimensionalized form. The vertical error bars represent the difference between the grayscale interpolation results for $h_{min}$ for the two individual imaging wavelengths.

We performed measurements on clean glass and partially wetting PC substrates, but no significant difference in the minimum film thickness $h_{min}$ on the two substrates was detected. For unstable rupturing films on partially wetting PC, we measured the value of $h_{min}$ by considering a cross-section as indicated in Fig. 2a in the undisturbed film between two dry spots. In the case of films thinner than 40 nm, the density of dry-spots was too high to find such a location and only wetting substrates were used to determine $h_{min}$. The numerical and experimental data in Fig. 4a agree to within 20% and the trend of $h_{min}$ ($Re_0$) is well reproduced. For higher $Re_0$, the quantitative agreement is almost perfect, whereas the simulations for low $Re_0$ slightly underestimate the impact of the air-jet. In the case of $U_{sub} = 2$ mm/s, the relative error in the film thickness measurement becomes appreciable as a film thickness $h_{min} < 10$ nm is reached, which is the lower resolution limit of our interference technique.

The minimum film thickness decreases with increasing jet Reynolds number $Re_0$ and decreasing substrate velocity $U_{sub}$ and follows a power law of roughly $Re_0^{-2}$, as indicated by the dashed line. However, the actual value of the exponent in simulations and experiments is slightly lower and seems to decrease with increasing substrate velocity $U_{sub}$. The numerical and experimental results for glycerol, 1EG and H2O in Fig. 4b are similar to the data for 3EG, with the exception of the measurement data for $h_{min} \approx 100$ nm, where evaporation enhances the thinning of the liquid films of 1EG and water. Evaporative effects also cause the deviation from a straight line in Fig. 4c, where the curves for 1EG and H2O for small velocity $U_{sub}$ display a higher slope for $h_{min} < 100$ nm. In the regime where evaporation does not play a significant role, i.e. for larger $U_{sub}$, the slopes appear to depend slightly on the viscosity of the liquid. The measurement and simulation data for water follow a power law $h_{min} \sim U_{sub}^{-0.85}$. We non-dimensionalized $h_{min}$ with the capillary length $l_{cap} = \sqrt{\frac{\gamma}{\rho_{gas}}} = \frac{\sqrt{\gamma}}{\rho_{gas}}$, a material parameter that ranges between 2 and 2.7 mm for the liquids considered. Another possible choice would be to normalize $h_{min}$ with the initial film thickness $h_0$. However, we carried out experiments with initial film thickness $h_0 = 3.3, 5$ and 17 μm and found identical values for $h_{min}$. Similarly, in Ref. [13] we found that for appreciable deformations ($h_{min}/h_0 < 0.5$) the value of $h_{min}$ is essentially independent of $h_0$. We plotted all data in Fig. 4a–c versus the ratio of Weber number and capillary number $We/Ca$ in Fig. 4d. The Weber number is defined as

$$We_0 \equiv \frac{\rho_{gas} v^2 D}{\gamma},$$

i.e. the ratio between the dynamic pressure from the air-jet $\frac{1}{2} \rho_{gas} v^2$ and the capillary pressure scale $\gamma/(2D)$ of the liquid film, with values ranging from 0.4 to about 100 in our experiments. The power law $h_{min} \sim Re_0^{\frac{1}{2}}$ in Fig. 4a and b suggests a linear dependence on $We_0$. The capillary number $Ca$ corresponds to a dimensionless substrate speed $U_{sub}$

$$Ca \equiv \frac{\mu_{liq} U_{sub}}{\gamma}.$$
The capillary number ranges from $2 \times 10^{-3}$ for 1EG to 0.25 for glycerol in our experiments. Due to the small spread in the power law exponents discussed above as well as the evaporation effects, the data do not collapse on a single line, but rather form a cloud of datapoints around a line corresponding to a power-law of approximately $(\text{We/Ca})^{-0.9}$.

Fig. 5a shows the minimum Reynolds number $R_{D,\text{min}}$ needed to obtain a certain minimum film thickness $h_{\text{min}}$ in a liquid film of 3EG with an initial thickness $h_0 = 5 \mu m$ as a function of capillary number $Ca$. This critical Reynolds number scales as $R_{D,\text{min}} \sim Ca^{0.52}$ for $h_{\text{min}} = 500$ nm with a slightly increasing exponent for lower $h_{\text{min}}$. Since the jet pressure distribution $P_{\text{jet}}$ decays much faster [37-40,13] with distance from the nozzle than the shear stress distribution $\tau_{\text{jet}}$, the two relevant components of $Q$, at the front rim are $\tau_{\text{jet}} h_{\text{cap}}^2/(2 \mu)$ and $U_{\text{sub}} h_{\text{min}}$. From a balance of these terms and the approximate scaling $\tau \sim \dot{\gamma} = \text{We}$ [13], the critical Reynolds number required to reach a certain minimum film thickness scales with $\sqrt{U_{\text{sub}}}$ or $Ca^{0.5}$ in excellent agreement with Fig. 5a. For the same reason, the minimum film thickness $h_{\text{min}}$ scales with $\tau \sim \text{Re}_{\text{D}}$ as we Fig. 4a and b and $h_{\text{min}} \sim \text{Re}_{\text{D}} \sim \text{Ca}$ in Fig. 4c.

The dependence of $h_{\text{min}}$ for 3EG on the normalized stand-off distance $H/D$ between needle and thin film is shown in Fig. 5b for $\text{Re}_{\text{D}} = 730$ and $U_{\text{sub}} = 2$ mm/s. For small values of $H/D \leq 5$, $h_{\text{min}}$ is essentially independent of $H/D$. At higher distances $H/D > 10$, the minimum film thickness begins to increase considerably. Such a transition was also observed for stationary high velocity gas jets impinging onto thicker water layers [41,42]. For all experiments in Fig. 4 and in the remainder of this manuscript, we used a value of $H/D = 5$.

4.2. Track width $w_{\text{max}}$

We measured the maximum track width $w_{\text{max}}$ according to the definition introduced in Fig. 2a. In Fig. 6a we plot $w_{\text{max}}$ as a function of $\text{Re}_{\text{D}}$ for different values of $U_{\text{sub}}$, whereas Fig. 6b shows the same data, normalized by the capillary length $l_{\text{cap}}$ and plotted versus $\text{We/Ca}$. The symbols represent experimental data and the solid lines indicate the results from numerical simulations. Fig. 6c shows the numerically determined $w_{\text{max}}/l_{\text{cap}}$ as a function of $Ca$ for different values of $\text{Re}_{\text{D}}$. Especially for higher $\text{Re}_{\text{D}}$, the experimentally found $w_{\text{max}}$ is well reproduced by the simulations. The normalized track width $w_{\text{max}}/l_{\text{cap}}$ follows a power law of $(\text{We/Ca})^{-0.37}$ for $\text{Re}_{\text{D}}$ with exponent $0.37$. The power law dependence on $Ca$ in Fig. 6c changes with increasing $\text{Re}_{\text{D}}$ from $w_{\text{max}} \sim Ca^{0.37}$ for $\text{Re}_{\text{D}} = 200$ to approximately $w_{\text{max}} \sim Ca^{0.3}$ for $\text{Re}_{\text{D}} = 1000$.

Fig. 7 shows interference microscopy images illustrating the deformation due to an air-jet impinging on moving films of 3EG on different substrate materials, and for different values of $U_{\text{sub}}$ and $\text{Re}_{\text{D}}$. Fig. 7a and b were obtained at different combinations of $U_{\text{sub}}$ and $\text{Re}_{\text{D}}$, but exhibit similar shape and minimum film thickness. Fig. 7c-e and the video in the Supplementary Content were obtained on partially wetting polycarbonate substrates, which resulted in the appearance of dry-spots and dewetting. The dotted yellow lines represent the rim profiles extracted from the numerical simulations of a wetting film on a linearly translating substrate. The numerical results fit the experimental rim shape very well, except for the positions far away from the needle impingement zone, where the influence of the rotation of the substrate in experiments deviates from the linear motion assumed in the simulations.

4.3. Centerline height profile

Fig. 8a and b presents numerical simulations of the centerline height profile $h(x=0,y)$ for a thin film of 3EG with $h_0 = 5 \mu m$. In
Fig. 7. Air-jet induced tracks in 3EG films (a) on glass with \( U_{\text{sub}} = 2 \text{ mm/s} \), \( R_{\text{DP}} = 163 \) resulting in \( h_{\text{max}} = (505 \pm 6) \text{ nm} \), (b) on glass with \( U_{\text{sub}} = 16 \text{ mm/s} \), \( R_{\text{DP}} = 573 \) resulting in \( h_{\text{max}} = (450 \pm 13) \text{ nm} \), (c) on PC with \( U_{\text{sub}} = 8 \text{ mm/s} \), \( R_{\text{DP}} = 990 \) resulting in \( h_{\text{max}} = (126 \pm 2) \text{ nm} \) and \( N_{\text{fr}} = (3.2 \pm 3.0) \text{ mm}^{-2} \), (d) on PC with \( U_{\text{sub}} = 8 \text{ mm/s} \), \( R_{\text{DP}} = 1840 \) resulting in \( h_{\text{max}} = 42 \pm 10 \text{ nm} \) and \( N_{\text{fr}} = 24.7 \pm 5.5 \text{ mm}^{-2} \), and (e) on PC with \( U_{\text{sub}} = 2 \text{ mm/s} \), \( R_{\text{DP}} = 1131 \) resulting in \( h_{\text{max}} = (24 \pm 10) \text{ nm} \) (interpolated from measurements on a wetting substrate) and \( N_{\text{fr}} = (223 \pm 28) \text{ mm}^{-2} \). The dotted lines indicate the rim profiles from the numerical simulations for a wetting film. The dashed lines indicate the regions used for the data analysis presented in Fig. 9.

Fig. 8a, the substrate speed \( U_{\text{sub}} \) is varied for constant \( R_{\text{DP}} \). Fig. 8b illustrates the influence of \( R_{\text{DP}} \) for constant \( U_{\text{sub}} = 16 \text{ mm/s} \). Fig. 8c shows an experimental example for \( U_{\text{sub}} = 16 \text{ mm/s} \) and \( R_{\text{DP}} = 1770 \). With increasing \( U_{\text{sub}} \) the distance \( a_{\text{front}} \) from the impingement point \( y = 0 \) to the maximum of the front rim decreases and the apex height of the rim \( h_{\text{max}} \) generally decreases. However, around \( U_{\text{sub}} = 64 \text{ mm/s} \), the rim height develops non-monotonically [see Fig. 8a]. Higher values of \( R_{\text{DP}} \) increase \( a_{\text{front}} \) and \( h_{\text{max}} \) [see Fig. 8b]. An interesting feature in the profile at the highest value of \( R_{\text{DP}} \) is the appearance of ‘ripples’ in the incline between the origin and the rim, as indicated by arrows in Fig. 8b.

Fig. 8. (a and b) Numerical simulations of the steady-state centerline height profiles \( h(x = 0, y) \) for a film of 3EG with \( h_0 = 5 \mu m \) for (a) \( R_{\text{DP}} = 998 \) and different values of \( U_{\text{sub}} \), and (b) \( U_{\text{sub}} = 16 \text{ mm/s} \) and different values of \( R_{\text{DP}} \). (c) An experimental micrograph for 3EG on PC with \( R_{\text{DP}} = 1770 \) and \( U_{\text{sub}} = 16 \text{ mm/s} \) exhibits similar ‘ripples’ as in (b). The arrowheads in (b and c) indicate the positions of these ripples. The dashed yellow line corresponds to the jet trajectory along which the profiles of (a) and (b) were extracted.

4.4. Scaling analysis

Akhatov [43], Tsukker [44] and Glauert [37] derived a self-similar analytical solution for the wall-jet region [38] of a jet impinging on a solid wall. In the axisymmetric case [44,37], the wall shear stress at large distances from the impingement point scales as \( \tau_{\text{jet}} \sim r^{-11/4} \), whereas the stagnation pressure distribution \( P_{\text{jet}}(r) \) decays rapidly [39,13] for distances exceeding \( D \). We first consider the steady state height profile along the jet trajectory, i.e. the \( y \)-axis in our rectilinear model, corresponding to the profiles in Fig. 8a and b. For symmetry reasons \( \partial Q_{x}/\partial x = 0 \) and Eq. (1) reduces to \( \partial Q_{y}/\partial y = 0 \). For sufficiently large \( R_{\text{DP}} \) or sufficiently small \( U_{\text{sub}} \), the rim position \( y = -a_{\text{front}} \) by far exceeds the needle diameter \( D \). Consequently, the two dominant terms in Eq. (3) are the contributions from the jet shear and the motion of the substrate. The balance of these terms yields the scaling relation

\[
U_{\text{sub}} h_0 \sim \frac{h_0^2 \tau}{2 \mu} \sim \frac{h_0^2 \tau_{\text{max}}}{2 \mu a_{\text{front}}^{11/4}},
\]

resulting in

\[
a_{\text{front}} \sim \left( \frac{h_0 \tau_{\text{max}}}{2 \mu U_{\text{sub}}} \right)^{4/11}.
\]

Our next step is the derivation of a scaling relation for the track width \( W_{\text{max}} \). We consider the region around the rim at the location \( y \approx 0 \). Eq. (1) reduces to

\[
\frac{\partial Q_{x}}{\partial x} + \frac{\partial Q_{y}}{\partial y} = 0.
\]

Again, the dominant terms in Eqs. (2) and (3) are those corresponding to shear stress and substrate motion. As we are interested in the rim profile, the relevant length scales for the \( x \)-and \( y \)-directions are \( W_{\text{max}} \) and \( a_{\text{front}} \), respectively. In the same manner as above, we arrive at the scaling relation

\[
U_{\text{sub}} h_0 \sim \frac{\tau_{\text{max}}}{W_{\text{max}}^{11/4}}.
\]

which is equivalent to

\[
W_{\text{max}} \sim \left( \frac{\tau_{\text{max}}}{U_{\text{sub}}^{3/2}} \right)^{4/11} \sim \left( \frac{\text{We}}{\text{Ca}} \right)^{4/11},
\]

in excellent agreement with the results in Fig. 6a–c.

4.5. Film rupture and dry-spot formation

The images in Fig. 7c–e show that the density of dry spots nucleated in liquid films on partially wetting substrates strongly increases with decreasing minimum film thickness \( h_{\text{min}} \). This is quantified in Fig. 9, where we plotted the density of dry spots, \( N_{\text{dp}} \) for 3EG and 1EG films on polycarbonate as a function of \( R_{\text{DP}} \), the ratio \( \text{We}/\text{Ca} \) and \( h_{\text{min}} \). The dry-spot density \( N_{\text{dp}} \) was measured by manual logging of each rupture site throughout the full duration of the experiment, i.e. during close to one full revolution of the substrate. This corresponds to a total track length of 3–6 cm, depend-
sites in both liquids is comparable. This may be explained by the fact that 1EG and 3EG are chemically very similar. Experiments with glycerol (data not shown) exhibited rupture events at higher minimum film thicknesses (\(h_{\text{min}} \approx 5 \text{ nm} \) at \(h_{\text{min}} = 300 \text{ nm} \) than 1EG and 3EG on PC. However, the experimentally accessible range in \(h_{\text{min}} \) was insufficient to perform systematic film rupture experiments with this liquid.

The data in Fig. 9c exhibit two regimes with approximate power law scaling \(N_{h} \sim h_{\text{min}}^{2} \) and \(N_{h} \sim h_{\text{min}} \). The transition between two regimes lies around \(h_{\text{min}} \approx 45 \text{ nm} \). The image in Fig. 7d illustrates a measurement close to that transition point. The power law \(N_{h} \sim h_{\text{min}}^{2} \) for low values of \(h_{\text{min}} \) is close to the experimental observation on thin films of polystyrene on silicon wafers [18–20] and is consistent with our earlier analysis of 3EG on PC (see Fig. 6d for \(R_{\text{D}} > 100 \) in Ref. [13]). For higher values of \(h_{\text{min}} \), a power law exponent of \(-2\) is more suitable. We attribute this transition to a confinement effect when the average distance between rupture sites becomes comparable to or larger than \(l_{1}\). This leads to an effectively one-dimensional film rupture geometry, which explains the reduction of the power law exponent [18]. If we assume that the disjoining pressure \(P_{\text{D}}\) is governed by a non-retarded van der Waals interactions, such that \(P_{\text{D}} = A/6\pi \eta r^{3} \), the density of holes in a flat “two-dimensional” film is related to the square of the most unstable wavenumber [45], \(k_{\text{max}}\) according to

\[
N_{h_{2D}} = \left( \frac{k_{\text{max}}}{2\pi} \right)^{2} = \left( \frac{1}{2\pi} \right)^{2} \left( \frac{h_{\text{min}}}{l_{1}} \right)^{2} = \frac{A_{\text{eff}}}{16\pi^{2}h_{\text{min}}^{2}}. \tag{18}
\]

In the one-dimensional regime this is

\[
N_{h_{1D}} = \frac{1}{l_{1}\eta} = \frac{k_{\text{max}}}{2\pi l_{1}} = \frac{1}{l_{1}h_{\text{min}}} \sqrt{\frac{A_{\text{eff}}}{16\pi^{2}h_{\text{min}}^{2}}}. \tag{19}
\]

where \(l_{1}\) is the average distance between dry-spots and \(l_{1}\) is the width of the region within which the dry-spots are counted [see Fig. 7c–e]. This distance \(l_{1}\) is approximately equal to \(D\) for \(h_{\text{min}} > 45 \text{ nm} \). Based on our data for \(h_{\text{min}} < 50 \text{ nm} \), we find an effective Hamaker constant \(A_{\text{eff}} = 2.3 \times 10^{-21} \text{ J} \). We also calculated the Hamaker constant using the Tabor–Winterton approximation [46–49]

\[
A_{\text{PC–3EG–air}} = \frac{3}{4} k_{\text{B}} T \left( \frac{\varepsilon_{\text{PC}} - \varepsilon_{\text{3EG}}}{\varepsilon_{\text{PC}} + \varepsilon_{\text{3EG}}} \left( \varepsilon_{\text{air}} - \varepsilon_{\text{3EG}} \right) \right) + \frac{3\eta_{\text{air}} v_{\text{e}}}{8\sqrt{2}} \times \left( \frac{n_{\text{air}}^{2} - 2n_{\text{air}} + n_{\text{air}}^{2}}{n_{\text{PC}}^{2} + n_{\text{3EG}}^{2}} \right) \left( 1 + \frac{R(h)}{n_{\text{air}}^{2} + n_{\text{PC}}^{2} + n_{\text{3EG}}^{2}} \right). \tag{20}
\]

where \(R(h)\) is a phenomenological factor accounting for retardation effects and \(\eta_{\text{air}} = 6.62 \times 10^{-13} \text{ J s} \) is Planck’s constant, \(k_{\text{B}} = 1.38 \times 10^{-23} \text{ J K}^{-1}\) Boltzmann’s constant and the resonance frequency \(v_{\text{e}} = 3 \times 10^{15} \text{ Hz} \). Using the values for \(n\) and \(\varepsilon\) from Table 1, \(\varepsilon_{\text{PC}} = 2.9\) and \(\varepsilon_{\text{3EG}} = 1.58\), we find \(A_{\text{PC–3EG–air}} = 2.18 \times 10^{-21} \text{ J} \). The retardation effect becomes important and \(R(h)\) goes to zero for \(h > 5–10 \text{ nm} \), such that for film thicknesses in the range of our experiments, the attractive Keesom and Debye interactions [the first term in Eq. (20)] dominate over the repulsive London interactions [the second term in Eq. (20)] [47–49]. A value \(A_{\text{PC–3EG–air}} = 2.18 \times 10^{-21} \text{ J} \) is found, which is very close to the observed value.

### 4.6. Residual droplet distribution

Immediately after the film rupture experiments described above, the patterns of residual droplets were recorded using a microscope. Examples of typical microscopy images are shown in Fig. 10. With decreasing residual film thickness, the droplet distributions change from stripes of droplets aligned perpendicular to...
the direction of motion [Fig. 10a], through a polygonal pattern of droplets [Fig. 10b] to a more dense and irregular distribution of many small droplets [Fig. 10c]. This trend reflects the transition in the dependence of dry-spot density $N_d$ on minimum film thickness $h_{\text{min}}$.

Using the image processing toolbox of MATLAB, we extracted the droplet density $N_d$ and average droplet radius $<R_d>$ from a random selection of 10 microscope images for each experiment. In Fig. 11a, histograms of droplet radii are shown for the measurements corresponding to Figs. 7c–e and 10. The histograms use 30 logarithmically spaced bins and are normalized with respect to the total number of droplets. The average droplet radius $<R_d>$ and corresponding error bars were based on the average value and standard deviation between the selected images. Fig. 11b presents $N_d$ and $<R_d>$ as a function of minimum film thickness $h_{\text{min}}$. The droplet density $N_d$ was determined in a manually selected region of interest of width $l_3$, illustrated by the green lines in Fig. 10. Fig. 11a and the blue symbols in Fig. 11b show that for a higher $N_d$, the uniformity of $<R_d>$ increases, i.e. the histograms become narrower and the average radius better defined. Lower droplet densities, like the one illustrated in Fig. 10a, consist of much fewer and bigger droplets as well as small satellite droplets between them. This renders the histograms wider and thus the standard deviations larger.

To illustrate the growth dynamics of the dry-spots and the formation of residual droplets, we present an image sequence from a measurement with 3EG on PC. Three nucleated dry-spots grow with an initial dewetting speed of approximately 850 $\mu$m/s. The dewetting rims of the receding contact lines are corrugated and exhibit a well-known rim instability [50–53]. The fastest growing wavelength $k_{\text{max}}$ for this instability was found to depend on the width $b$ of the rim and the contact angle $\theta$, according to $k_{\text{max}}/b = \beta(\theta)$. Brochard et al. [50] report $\beta = 4.2$ and Münch and Wagner [51,53] found $\beta = 2.4$ to 3.2.

![Fig. 10. Examples of droplet patterns resulting from film rupture and dewetting of 3EG films, corresponding to Fig. 7c–e, respectively. (a) $h_{\text{min}} = (126 \pm 2)$ nm: $N_d = (19 \pm 13)$ mm$^{-2}$ and $<R_d> = (13 \pm 5)$ $\mu$m; (b) $h_{\text{min}} = (42 \pm 10)$ nm: $N_d = (145 \pm 48)$ mm$^{-2}$ and $<R_d> = (6 \pm 1)$ $\mu$m; (c) $h_{\text{min}} = (24 \pm 10)$ nm: $N_d = (1766 \pm 300)$ mm$^{-2}$ and $<R_d> = (3 \pm 0.3)$ $\mu$m. The red dashed circles indicated the needle diameter, the green solid lines enclose the area used for calculation of droplet densities. The blue dotted line indicates the jet trajectory. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)](image)

![Fig. 11. (a) Histograms of droplet radii extracted from the experiments illustrated in Figs. 7c–e and 10. (b) Dependence of residual droplet density $N_d$ and average droplet radius $<R_d>$ on $h_{\text{min}}$ for 3EG films on PC and two different values of $U_{\text{sub}}$. The solid and dashed lines represent the theoretical scaling relations derived in Section 4.7.](image)

![Fig. 12. Microscope images at different points in time illustrating the growth of dry-spots, the rim instability in the receding contact line and creation of droplets where the dry spots coalesce.](image)

4.7. ‘Geometrical’ estimate of droplet density

The images in Fig. 12 clearly show how bulges of the rim instability can turn into residual droplets upon rim coalescence. In this section we present two primarily geometric models for the density

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of residual droplets $N_d$ for both the one- and two-dimensional regime.

4.7.1. Two-dimensional regime

An estimate for the total areal density of droplets is given by

$$N_d = N_c + (n_1 - c_1)N_h.$$  \hspace{1cm} (21)

Here, $N_c$ is the areal density of nodes on the Voronoi grid [19,54] based on the centers of the dry-spots. These are illustrated in Fig. 13 as solid circles for two idealized, periodic arrangements of dry-spots, denoted by asterisks. The lines connecting the nodes in Fig. 13 are termed segments. The segments represent rivulets, resulting from the coalescence of two adjacent dewetting rims. Fig. 13d presents a sketch of a rivulet. The density of nodes is proportional to the density of dry spots, $N_c = c_2N_{h_0}$, where the numerical prefactor $c_2 = 1$ for a square lattice [see Fig. 13a] and $c_2 = 2$ for a hexagonal lattice of dry-spots [Fig. 13b]. The parameter $N_h$ represents the areal density of segments, which equals $N_h = c_3N_{h_0}$, with $c_3 = 2$ for the square geometry and $c_3 = 3$ for a hexagonal lattice. The number of droplets $n_1$ per segment depends on the corresponding rivulet width $b$ and the length of the segment $l_{2,3}$, i.e. the average distance between two nodes, as illustrated in Fig. 13d. The constant $c_1 \approx 2$ in Eq. (21) compensates for the effect that the outer droplets of each segment are merged with the corner droplets.

The segment length $l_2 = c_4/\sqrt{N_{h_0}}$, where $c_4 = 1$ for a square lattice and $c_4 = 6.52 \approx 0.62$ for a hexagonal pattern. An estimate for the volume of each rivulet segment is given by the accumulated volume $c_5h_{\text{max}}l_2^2$, as indicated by the purple regions in Fig. 13. Here, $c_5 = 0.5$ for the square geometry and $c_5 = 0.5\sqrt{3} \approx 0.866$ for a hexagonal lattice. This volume is distributed over a rivulet of length $l_2$, sketched in Fig. 13d. Approximating the cross-sectional area $A$ of the rivulet as a circle segment with contact angle $\theta$ gives

$$A = \frac{\theta - \sin \theta \cos \theta}{4 \sin^2 \theta} b^2 \equiv c_6b^2,$$  \hspace{1cm} (22)

with $c_6 = 0.091$ for $\theta = 30^\circ$. Equating $l_2A = c_5h_{\text{max}}l_2^2$ yields $b = \sqrt{h_{\text{min}}l_2}/(c_5/c_6)$. The number of droplets per segment thus becomes

$$n_x = \frac{l_2}{ld} = \frac{\sqrt{l_2}}{\beta \sqrt{\left(\frac{c_5}{c_6}\right)b_{\min}}} = \frac{N_d^{1/4}}{\beta \sqrt{\left(\frac{c_5}{c_6}\right)b_{\min}}}. $$  \hspace{1cm} (23)

Eq. (21) now yields

$$N_d = (c_2 - c_1c_3)N_h + \frac{c_2N_{h_0}^{1/4}}{\beta \sqrt{\left(\frac{c_5}{c_6}\right)b_{\min}}}. $$  \hspace{1cm} (24)

Using the scaling $N_d = c_7h_{\text{min}}^{0.9}$ found in Section 4.5 with $c_7 \approx 1 \times 10^{-22}$ m$^2$, which corresponds to a disjoining pressure $\Pi \sim h^{-3}$, we find

$$N_d = c_7(c_2 - c_1c_3)h_{\min}^{4/5} + \frac{c_1^{1/4}c_2h_{\min}^{3.5}}{\beta \sqrt{\left(\frac{c_5}{c_6}\right)b_{\min}}}. $$  \hspace{1cm} (25)

The two terms in Eq. (25) have power law dependencies $N_d \sim h_{\min}^{0.9}$ and $N_d \sim h_{\min}^{3.5}$. Using the constants $c_1 = 2$, $c_2$ to $c_5$ for a hexagonal lattice, $c_4$ corresponding to $\theta = 30^\circ$ and $c_7 = 1 \times 10^{-22}$ m$^2$ from the experimentally determined $N_d$ in Section 4.5 and $\beta = 3$, we plot Eq. (25) as the red solid line in Fig. 11b. The first term in Eq. (25) is generally negative, yielding an approximate power law $N_d \sim h_{\min}^{-0.1}$. The match with the experimental data in Fig. 11b is excellent.

The average droplet radius $\langle R_d \rangle$ corresponds to the base radius of a spherical cap of volume $\langle V_d \rangle$.

$$\langle R_d \rangle = \frac{\sin \theta}{\sqrt{\frac{3}{2} - 3\cos^2 \theta + \cos^2 \theta}} \cdot \sqrt{\langle V_d \rangle} \approx 1.32\sqrt{\langle V_d \rangle}, $$  \hspace{1cm} (26)

for $\theta = 30^\circ$. From the conservation of mass the average droplet volume $\langle V_d \rangle = h_{\min}/N_d$. We plotted the resulting expression for $\langle R_d \rangle$ as the blue solid line in Fig. 11b and again find an excellent match with the measurement data for low values of $h_{\min}$. The blue line follows an approximate scaling of $\langle R_d \rangle \sim h_{\min}^{1/4}$.

For comparison, previous investigations [18–20,27] reported similar exponents for polystyrene films on silicon: around $\langle R_d \rangle \sim h_{\min}^{0.4}$ and $N_d \sim h_{\min}^{-0.5}$ for $h_{\min} \leq 30$ nm, possibly decreasing to $N_d \sim h_{\min}^{1.5}$ for $h_{\min} > 30$ nm [27]. Sharma and Reiter [20] derived a power law $\langle R_d \rangle \sim h_{\min}$ based on a model of dry-spot coalescence and subsequent Rayleigh instability of the rim segments. They observed a power law $\langle R_d \rangle \sim h_{\min}$ for material systems where fingering instabilities contribute to the formation of residual droplets.

4.7.2. Quasi-one-dimensional regime

For large values of $h_{\min}$, the spacing $L_{st}$ between dry-spots increases and $N_d$ decreases. The rupture is basically a one-dimensional process with a different power law, which we found in Section 4.5 to be

$$N_d \sim k_{\text{max}} \sim h_{\text{min}}^{-2}. $$  \hspace{1cm} (27)

In this case, dry-spots are spaced with an average distance $L_{st}$ according to $N_d = 1/(L_{st}h_{\text{min}})$, where $L_{st}$ is the length of the valley around $h_{\text{min}}$, illustrated by the red dashed lines in Fig. 7. The droplets are distributed over a slightly wider area, corresponding to the breakup of rivulets with length $l_3$ positioned between two dry-spots. Fig. 13c illustrates this one-dimensional configuration. The length of the rivulets is of the order of the needle diameter, $l_3 = f_dD$. From our analysis of droplet patterns, we found that the value of $f_d$ is between 3 and 6 for $h_{\min} \geq 50$ nm and scales approximately linear with minimum film thickness according to

$$f_d(h_{\text{min}}) \approx 2.7 \times 10^7 m^{-1} \cdot h_{\text{min}} - 1.45. $$  \hspace{1cm} (28)
The rivulet consists of the accumulated volume \(L_{th}h_{min}/2\), indicated by the purple region in Fig. 13c. Using the cross-section of the rivulet \(A = C_0b^2 = L_{th}h_{min}/2\), we obtain for the rivulet width \(b = \sqrt{L_{th}h_{min}/(2C_0)}\). The number of droplets for each rivulet is

\[
n_d = \frac{n_l}{L_{th}} = \frac{L_{th}/C_0}{\beta \sqrt{L_{th}h_{min}}},
\]

and the density of droplets thus becomes

\[
n_d = \frac{n_l}{L_{th}} = \frac{2C_0}{\beta} l_1^{-1.5} h_{min}^{-1/2}.
\]

If we approximate the experimental data for \(h_{min} > 45\) nm in Section 4.5 with \(N_d = C_9h_{min}^{-2}\) where \(C_9 \approx 5 \times 10^{-8}\) m\(^2\), we obtain \(l_1 = h_{th}^{-1}/C_9 l_t\) for the one-dimensional case. Inserting in Eq. (30), gives

\[
n_d = \frac{C_9 \sqrt{2C_0} l_1}{\beta} l_1^{1.5} h_{min}^{-3.5}.
\]

Using \(l_1 \approx D\), we plot \(n_d\) as a red dashed line in Fig. 11b, and find an approximate power-law scaling \(N_d \sim h_{min}^{-3.5}\).

The volume in the rivulet segment of length \(l_1\) is accumulated from an area \(L_{th}/2\) with a thickness of \(h_{min}\) and thus following the reasoning of Eq. (26) for the two-dimensional case, we find that the average droplet radius in the one-dimensional region becomes

\[
\langle R_d \rangle \approx 1.32 \sqrt{\langle V_d \rangle} = 1.32 \frac{h_{min} l_t}{2n_l} = 1.32 \frac{\beta}{\sqrt{6} \sqrt{2C_0} l_t} h_{min}^{1.5}.
\]

Using \(l_1 \approx D\), we find a scaling \(\langle R_d \rangle \sim h_{min}^{1.5}\) plotted as the blue dashed line in Fig. 11b. The experimental data seems to lie lower than the model predictions for high values of \(h_{min}\) in Fig. 11b. The reason for this may be that the polydispersity of the experimental droplet sizes is not reflected in the geometric scaling analysis, where we calculated average values only. For low \(h_{min}\), the droplet size distribution is more narrow and is thus more correctly represented in the analysis. Moreover, the tracks are narrower for smaller values of \(R_{th}\) or higher values of \(U_{sub}\) corresponding to a higher lateral curvature \(\frac{\partial h}{\partial y}(y = 0)\) at the track center line. This may not be entirely consistent with our assumption of a uniform film thickness \(h = h_{min}\) in the region of interest \(|x| < l_1/2\), upon which Eqs. (27)–(32) are based.

\section{5. Conclusions}

We studied the deformation of thin liquid films induced by a circular laminar air-jet impinging at normal incidence and moving with respect to the substrate. Quantitative measurements of the thin film deformation for four different liquids were acquired by means of dual-wavelength interference microscopy, combined with a custom-designed optically-accessible spin-coater. Moreover, we developed a numerical model for the deformation of the liquid films, based on the lubrication approximation and coupled to calculations of the shear stress and pressure distribution of the impinging air-jet. Systematic experiments and numerical results were in excellent agreement. Moreover, we performed a comprehensive scaling analysis that reproduce the observed power law dependencies for the shape and residual film thickness of the liquid film deformation.

On partially wetting polycarbonate surfaces, we observed rupture and dewetting of the liquid films, a relevant phenomenon for coating technology \cite{14} and immersion lithography \cite{15–17}. We determined the density of dry spots as well as the density and size distribution of residual droplets as a function of the minimum film thickness. We found reproducible power laws, independent of the liquid viscosity, jet Reynolds number and substrate speed within experimental accuracy.

For low film thicknesses, we measured power laws for the dry-spot density very similar to those found by others for polystyrene melts on silicon wafers \cite{18–20,27}. For our material system of polar liquids on polycarbonate, the dry-spot density coincides with the most unstable wavelength predicted by linear stability analysis for a Van der Waals type disjoining pressure with a value of the Hamaker coefficient in agreement with the retarded version of the Tabor–Winterton approximation.

A transition in the power law for the density of dry-spots was observed at a minimum film thickness of approximately 45 nm as a consequence of lateral confinement of the area of low film thickness. The transition occurs approximately when the most unstable wavelength exceeds the base width of the track. In a future paper, we will further address confinement effects by presenting results obtained with wider, non-circular jets.

Finally, we developed a geometrical model for the residual droplet distribution corresponding to the observed dry-spot density. Our analysis is based on the observation that the rim instability of the dewetting dry-spots \cite{50,51} determines the final droplet pattern. We obtain a good match with the experimentally observed average droplet size and density, especially for low film thickness. In technological applications residual droplets are often undesirable, because their presence interferes with subsequent process steps, their evaporation induces temperature gradients and recondensation may occur on cooler components.

\section*{Acknowledgments}

This work is part of the research program ‘Contact Line Control during Wetting and Dewetting’ (CLC) of the ‘Stichting voor Fundamenteel Onderzoek der Materie (FOM)’, which is financially supported by the ‘Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO)’. The CLC program is co-financed by ASMIL and Océ.

\section*{Appendix A. Supplementary material}

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.jcis.2013.06.011.

\section*{References}