Controlled destabilization of liquid coatings on partially wetting substrates using laminar air-jets[1]

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Introduction

In immersion lithography [1], light is focused through a layer of water to achieve higher imaging resolution. At high scan speeds, thin liquid films are entrained on the partially wetting, photoresist-coated wafer. These films rupture, dewet and leave droplet patterns behind on the surface [2]. The rupture of thin liquid films is also a relevant phenomenon in discrete area coating, where air-jets are used to initialize liquid redistribution [3]. We are investigating film rupture and study techniques that can control and accelerate the rupture process.

Here we present experiments and numerical simulations of the dynamics of thin liquid films on partially wetting substrates subjected to impinging laminar air-jets [4]. We systematically studied the shape of the disturbed liquid film and rupture time as a function of the Reynolds number of the air-jet and the nozzle diameter experimentally and numerically. We are currently extending our research towards moving air-jets. An impression of the first results is given at the end of this extended abstract.

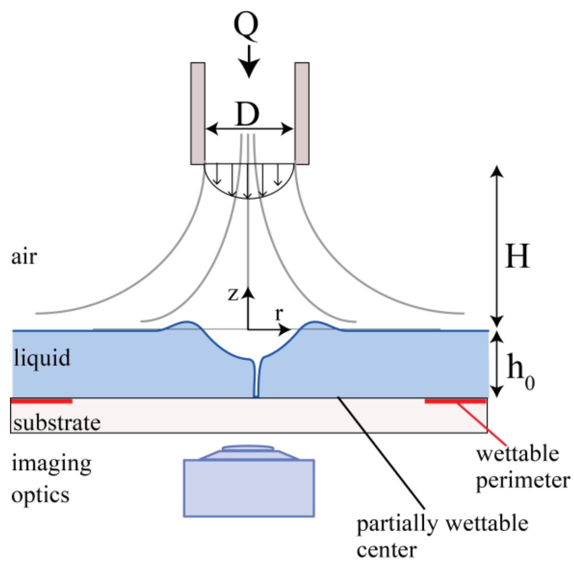

Figure 1: *Schematic representation of the experimental configuration.*

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Experimental procedure

For our experiments we used transparent plates of polycarbonate (PC, Sabic, Lexan, thickness 1mm), of which we rendered the perimeter wettable using UV/Ozone treatment and a shadow mask to protect the partially wetting center area. A liquid film of triethylene glycol (3EG, Sigma, Purity ≥99%, receding contact angle θ_{rec}=29° on PC) with a thickness h_0=5μm was spin coated onto the substrate. The liquid film was subjected to a laminar jet of clean air from a hollow needle with diameter D, positioned at a height H=2mm above the film. Reflection interference micrographs were recorded from below using a CCD camera (Guppy, Allied Vision Technologies) and an imaging system (InfiniTube) using LED illumination (λ≈650nm) [Figure 1], which allow for reconstruction of the time dependent film shape. Experiments with moving substrates were performed by integrating a turning table in the setup described above.

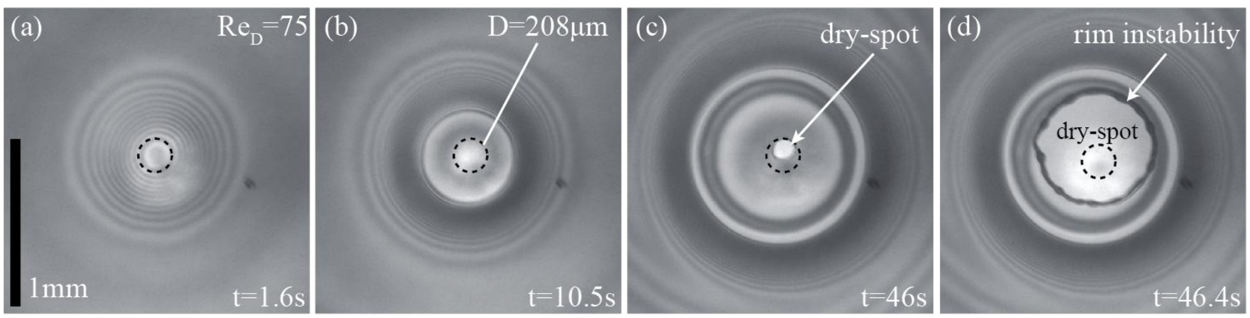

Figure 2: *Example of an experiment at Re_D=75. The air-jet is switched on at t=0, rupture occurs at t=45.9s.The dashed circle indicates the inner diameter of the hollow needle (D=208μm).*

Numerical models

To evaluate the influence of an air-jet on the thin liquid film numerically we modeled the impinging jet using the stationary incompressible Navier Stokes equation. The pressure and shear stress distributions at the location of the liquid film were implemented in a numerical model for thin film flow, based on the lubrication approximation [5]. The surface forces that play a role in film rupture, formation of a three phase contact line and dewetting are represented in a so-called disjoining pressure. The strength of this disjoining pressure is strongly dependent on the local film thickness. In our model, we implemented the empirical expression for the disjoining pressure Π(h) as introduced by Schwartz and Eley [6], including contributions of short range repulsion and long range attraction:

$$\Pi(h) = \gamma(1-\cos\theta)\frac{(n-1)(m-1)}{(n-m)h^*}\left[\left(\frac{h^*}{h}\right)^n - \left(\frac{h^*}{h}\right)^m\right] \qquad \text{(Equation 1)}$$

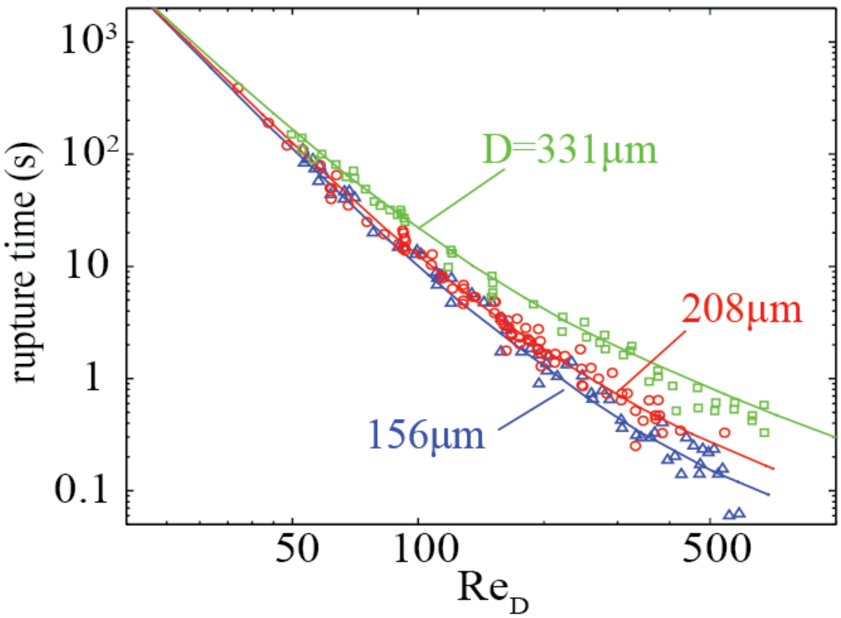

Figure 3: *Experiments (symbols) and simulations (solid lines) of film rupture time as a function of jet diameter D and Reynolds number Re_D.*

Results and discussion

Figure 2 shows the local thinning of a thin film by means of an impinging air-jet and the nucleation of a dry-spot. Figure 2(d) illustrates a hydrophdynamic instability of the dewetting rim [7], which causes the slightly irregular morphology of the perimeter of the growing dry-spot.

The time between switching on the air-jet and film rupture is plotted as a function of jet Reynolds number Re_D in Figure 3. The symbols represent the experimental data, the solid lines the results of the numerical simulations. The rupture time varies from more than 100s to less than 100ms. Narrower jets cause earlier rupture than wider jets. We systematically varied the disjoining pressure parameters h^*,m and n in Equation (1). Excellent agreement between experiments and simulations was found for h^*=10nm, m=4 and n=10. We used γ=45.5 mN/m as the surface tension of 3EG and $\theta = \theta_{rec}$=29° for the contact angle.

Moving jets

In the presence of relative motion between the sample and the jet, an elongated depression is created in the liquid film. Figure 4 shows an example of such an experiment and simulation. At high substrate velocity or low Re_D, the film thickness reduction is too small for film rupture to occur. We are currently investigating film rupture in this system more systematically.

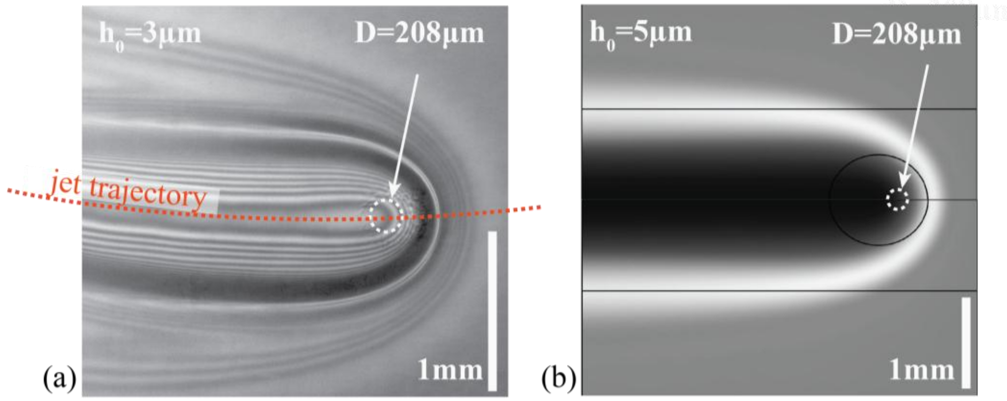

Figure 4: *Example of an air-jet impinging on a moving sample. The inner diameter of the hollow needle (D=208μm) is illustrated by the dashed circles. (a) Experimental interference micrograph for a circular trajectory. The initial film thickness h_0= 3 μm. The substrate velocity U≈2mm/s and Re_D=500 (b) Simulation of a film with thickness h_0= 5 μm, U=2mm/s and Re_D=503.*

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References

[1] R.H. French and H.V. Tran, *Annu. Rev. Mater. Sci.* **39**, 93 (2009).

[2] G. Reiter, *Phys. Rev. Lett.* **68**, 75 (1992).

[3] C.L. Bower, E.A. Simister, E. Bonnist, K. Paul, N. Pightling and T.D. Blake, *AIChE J.* **53**, 1644 (2007).

[4] C.W.J. Berendsen, J.C.H. Zeegers, G.C.F.L. Kruis, M. Riepen and A. A. Darhuber, *Langmuir*, doi: 10.1021/la301353f (2012).

[5] A. Oron, S.H. Davis and S.G. Bankoff, *Rev. Mod. Phys.* **69**, 931 (1997).

[6] L.W. Schwartz and R.R. Eley, *J. Colloid Interface Sci.* **202**, 173 (1998).

[7] F. Brochard-Wyart and C. Redon, *Langmuir* **8**, 2324 (1992).