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Infrared laser induced rupture of thin liquid films on stationary substrates

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We studied the deformation and destabilization of thin liquid films on stationary substrates via infrared illumination. The film thickness evolution was measured using interference microscopy. We developed numerical models for the temperature evolution and the liquid redistribution. The substrate wettability is explicitly accounted for via a phenomenological expression for the disjoining pressure. We systematically measured the film thinning- and rupture dynamics as a function of laser power, which are accurately reproduced by the simulations. While smaller laser spots generally lead to shorter rupture times, the latter can become independent of the spotsize for very narrow beams due to capillary suppression. © 2014 AIP Publishing LLC.

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In immersion lithography,^{1,2} the optical resolution is enhanced by introducing a water layer between the objective lens and the wafer to be exposed. In order to maximize throughput, a high relative speed between the wafer and the projection optics is desirable. Above a critical velocity, however, a thin layer of water is entrained on the photoresist surface, which subsequently breaks up into droplets. The motivation for this study is to evaluate infrared irradiation as a potential mechanism for controlling the rupture process by destabilizing the liquid film as swiftly as possible. Controlled dry-spot formation is also applied to initiate the redistribution of liquid films coated on chemically patterned surfaces.³

The deformation of liquid films by means of non-uniform temperature distributions has been studied extensively.^{4–8} The rupture of liquid films subjected to thermal gradients^{9–14} was studied in the context of film cooling, where dryout decreases the effectiveness of the heat removal. Recently, Singer *et al.* used a laser-based technique to induce localized dewetting of thin polymer films.¹⁵

In this article, we present quantitative experiments and numerical simulations of the deformation and rupture of non-volatile liquid films by non-uniform temperature distributions, induced by infrared (IR) laser heating of the substrate. We studied the dependence of the rupture time of partially wetting thin films on the two most important experimental parameters, i.e., the laser power and the laser spot size. Figure 1(a) shows a sketch of the experimental setup. We use cylindrical coordinates (r, z) , where r is the radial coordinate. The line $r = 0$ (i.e., the z -axis) coincides with the vertically oriented optical axis, which is normal to the horizontal substrate surface. The plane $z = 0$ corresponds to the focal plane, which was adjusted to coincide with the top surface of the substrate. The optically thin polycarbonate (PC) substrate is transparent to visible light and partially absorbs the IR light (absorption coefficient $\alpha_{PC} = (30 \pm 1) \text{ m}^{-1}$ at 1470 nm). A liquid film was deposited on the top side of the substrate by spin coating. The resulting initial film thickness

h_0 in the order of several microns was measured with a spectral interference technique.¹⁶ The deformation and rupture of the liquid film was monitored *in-situ* using dual wavelength interference microscopy.¹⁷

The light source is a multimode diode laser system (Lumics LU1470C20-C) with a center wavelength of $\lambda = 1470 \text{ nm}$ and a maximum output power of 20 W. The diverging laser light exiting from the fiber output (diameter $400 \mu\text{m}$) is refocused to a spot in the order of $200 \mu\text{m}$. We adjusted the laser power range via the current to the laser diode. Linear variation of power within the selected range was achieved by modulating the laser at a frequency of 50 Hz using a variable duty cycle. We characterized the intensity profile $I(r, z)$ of the laser beam using a home-built scanning slit setup. We represent $I(r, z)$ by a Gaussian distribution of variable width $w(z) = w_0 + a|z|$

FIG. 1. (a) Sketch of the experimental setup for studying the laser-induced deformation of thin liquid films on stationary substrates. (b)–(d) Laser-induced rupture of a thin film of 3EG on a PC substrate for $P = 120 \text{ mW}$, $d_{\text{sub}} = 750 \mu\text{m}$ and $h_0 = 5 \mu\text{m}$.

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$$I(r, z) = \frac{2P}{\pi[w(z)]^2} \exp\left[-\frac{2r^2}{[w(z)]^2}\right] \exp(-\alpha_{PC}|z|), \quad (1)$$

where P is the selected laser power. We determined the waist radius as $w_0 = 120 \mu\text{m}$ and $a = 0.3$. We used the completely wetting liquid squalane and the partially wetting tri(ethylene glycol) (3EG). Their surface tension $\gamma(T)$, viscosity $\mu(T)$, density $\rho(T)$, as well as the thermal properties of PC are given in the supplementary material.¹⁸

In order to simulate the thermocapillary deformation of thin liquid films, two coupled models are required: one for the time-dependent temperature distribution¹⁸ and one for the flow of the liquid film, both of which were implemented using the finite-element software Comsol 3.5a.

Figure 2(a) shows simulated surface temperature profiles, $\Delta T(r, t) \equiv T(r, z = 0, t) - T_0$ at different times t for a laser spotsize of $w_0 = 120 \mu\text{m}$. Here, $T_0 = 293 \text{ K}$ is the uniform initial temperature. The maximum temperature, which occurs at $r = 0$, rapidly increases in time after the laser is switched on at $t = 0$ and approaches a steady-state value after approximately 10 s. Due to heat conduction in the radial direction, the temperature rise extends further than the illuminated region ($r \lesssim w_0$). Figure 2(c) shows the radial derivative of the surface temperature profile, $\frac{\partial T}{\partial r}(r, z = 0, t)$. Its maximum absolute value is located off-center and reaches a steady-state much faster, with a risetime of approximately 0.06 s. Figures 2(b) and 2(d) show the maximum surface temperature rise and maximum absolute value of the radial temperature gradient in steady state as a function of P . Both quantities increase linearly with P .

Since the initial film thickness $h_0 = 5 \mu\text{m}$ is much smaller than the diameter of the laser spot, we use the lubrication equation¹⁹ for describing the axisymmetric evolution of the liquid film thickness $h(r, t)$

$$\frac{\partial h}{\partial t} + \frac{1}{r} \frac{\partial}{\partial r} \left[r \left(\frac{h^2}{2\mu(T)} \tau_r - \frac{h^3}{3\mu(T)} \frac{\partial \mathcal{P}}{\partial r} \right) \right] = 0. \quad (2)$$

The augmented pressure

$$\mathcal{P} \equiv -\frac{\gamma(T)}{r} \frac{\partial}{\partial r} \left(r \frac{\partial h}{\partial r} \right) + \rho(T)gh - \Pi(h) \quad (3)$$

represents the influence of capillary pressure, hydrostatic pressure, and the disjoining pressure²⁰ Π . For the simulation of partially wetting liquids, we use a phenomenological equation for the disjoining pressure²¹

$$\Pi(h) = \gamma(1 - \cos \theta) \frac{(n-1)(m-1)}{(n-m)h^*} \times \left[\left(\frac{h^*}{h} \right)^n - \left(\frac{h^*}{h} \right)^m \right]. \quad (4)$$

For a completely wetting liquid, we set $\Pi = 0$.

Temperature gradients induce non-uniformities in the surface tension that redistribute the liquid away from the heated region. The term $\tau_r \equiv \frac{\partial \gamma}{\partial r} = \frac{\partial \gamma}{\partial T} \frac{\partial T}{\partial r}$ in Eq. (2) represents the thermocapillary shear stress acting on the thin

FIG. 2. Numerical results from heat transfer model for $P = 0.5 \text{ W}$, $w_0 = 120 \mu\text{m}$, and a substrate thickness $d_{\text{sub}} = 750 \mu\text{m}$. (a) Surface temperature profiles $\Delta T(r, z = 0, t)$. (b) Maximum temperature rise in steady-state ΔT_{max} as a function of P . (c) Radial surface temperature gradient $\frac{\partial T}{\partial r}(r, z = 0, t)$. (d) Maximum absolute value of temperature gradient in steady-state $|\frac{\partial T}{\partial r}|_{\text{max}}$ as a function of P .

film, where $T_s(r)$ is the temperature of the liquid-air interface. Due to the small film thickness and the absence of significant vertical temperature gradients, T_s is to good approximation equal to the substrate surface temperature $T_s \approx T(r, z = 0, t)$, which we obtain from the heat transfer calculations. The factor $\frac{\partial \gamma}{\partial T}$ quantifies the sensitivity of the surface tension to temperature variations. The boundary conditions represent axisymmetry at $r = 0$ and an undisturbed film at a distance $R_d = 2 \text{ cm}$ that is chosen sufficiently large such that the deformation does not reach the end of the domain. The initial condition is a film of uniform thickness $h(r, t = 0) = h_0$.

Figures 3(a) and 3(b) show interference micrographs of a thin squalane film at different times after switching on the laser beam. As a consequence of thermocapillary shear, a depression is formed in the liquid film in the vicinity of the laser spot. This “dip” is surrounded by a ring of increased film thickness, which we term “rim” [see Fig. 3(b)]. The deformation is nicely axisymmetric and deepens and widens in time. Figure 3(c) shows the minimum film thickness $h_{\text{min}}(t) = h(0, t)$ for $h_0 = 5.5 \mu\text{m}$ and different values of P . The solid lines are obtained from the numerical simulations, the symbols from the experiments, which agree very well. Higher laser powers result in a lower film thickness. Figure 3(c) also shows that after a certain time, the minimum film thickness scales as $h_{\text{min}} \sim t^{-1}$ (dashed line). An analogous result was observed by Darhuber and Troian,²² who derived an asymptotic solution of Eq. (2) in the form

FIG. 3. (a) and (b) Interference micrographs illustrating the thermocapillary deformation of a thin squalane film 2 and 8 s after switching on the laser beam at $t=0$ ($P=0.3$ W, $h_0=5.5$ μm , substrate thickness $d_{\text{sub}}=175$ μm). (c) and (d) Minimum film thickness $h_{\text{min}}(t)$ for (c) $h_0=5.5$ μm and different values of P and (d) $P=55$ mW and different values of h_0 . Solid lines represent numerical simulations, symbols experimental data. The dashed line in (d) corresponds to Eq. (5).

$$h_{\text{min}}(t) = \frac{2\mu(t-t_0)^{-1}}{\frac{\partial \gamma}{\partial T} \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial T}{\partial r} \right) \right]_{r=0}}, \quad (5)$$

where t_0 is an integration constant. This equation is valid for sufficiently small temperature variations such that μ can be considered temperature independent.

Figure 3(d) shows $h_{\text{min}}(t)$ for $P=55$ mW and different values of h_0 . Both the experiments and the numerical simulations show that h_{min} is independent of h_0 after approximately 10 s. The black dashed line in Fig. 3(d) represents the analytical solution of Eq. (5).

A thin liquid film on a partially wetting substrate is unstable to dry-spot formation and will eventually dewet the substrate. We studied the laser-induced rupture of partially wetting 3EG films on PC substrates. Figures 1(b)–1(d) show an example for $P=120$ mW and $h_0=5$ μm . Figure 1(b) shows a dry spot in the center of the deformation zone just after the moment of break-up. Figures 1(c) and 1(d) illustrate its expansion. Figure 4(a) presents the rupture time t_r , i.e., the time delay between switching on the IR-laser and film break-up, as a function of P . The red open circles represent experimental data. The solid blue line represents numerical simulations using the values $n=10$, $m=4$, $h^*=10$ nm, and $\theta=29^\circ$ in Eq. (4). These parameters are identical to those determined independently in Ref. 16. The match between experimental and numerical data is excellent.

The green squares, which are very well approximated by a power law $t_r \sim P^{-0.86}$ (green dashed line), correspond to numerical simulations assuming temperature-independent

FIG. 4. (a) Experiments (red open circles) and numerical simulations (solid blue diamonds and green squares) of the rupture time t_r as a function of P for $w_0=120$ μm , $h_0=5.5$ μm and $d_{\text{sub}}=750$ μm . (b) Numerical simulations of t_r as a function of w_0 for three values of P .

material parameters. The difference between the green line and the experimental data illustrates primarily the temperature dependence of μ , which reduces t_r tenfold at the highest power considered. The light-blue dashed and orange solid horizontal lines correspond to the rise times of the center temperature and the maximum temperature gradient. Figure 4(b) shows t_r as a function of w_0 for different values of P . The dashed line corresponds to a power law $t_r \sim w_0^{1.86}$. A narrower laser spot, thus, significantly accelerates the rupture process.

Film rupture is the terminal event of the deformation process. Thus, t_r is sensitive to the timescales of the temperature and the temperature gradient and the capillary transition time t_0 in Eq. (5). For sufficiently high laser powers, the latter can be disregarded. For the experiments in Fig. 4(a), the risetime of the temperature gradient is typically much smaller than t_r , such that we can assume the thermocapillary stress to be fully developed during the entire set of experiments. The disjoining pressure becomes important at very low values of h , where the capillary pressure gradients have become negligible, at least for sufficiently high laser powers. The rupture thickness h_r is then characterized by a balance of the flux terms corresponding to thermocapillary shear and disjoining pressure gradients in Eq. (2). For sufficiently high P , the width of the depression is larger than the maximally unstable wavelength of the spinodal instability²³

$$\lambda_{\max} = (8\pi^2\gamma)^{1/2} \left(-\rho g + \frac{\partial \Pi}{\partial h} \right)^{-1/2} \sim h^{5/2}. \quad (6)$$

Assuming w_0 and $\lambda_{\max}$ to be the proper length scales for $\partial\gamma/\partial x$ and $\partial\Pi/\partial x$, respectively, we find the scaling

$$h_r \sim [\Delta\gamma/(\gamma w_0)]^{-2/11} \sim P^{-2/11}, \quad (7)$$

which suggests that h_r is only very weakly dependent on P . A corresponding rupture time-scale follows from Eq. (2), considering thermocapillary shear as the main mechanism driving the film thinning, as

$$t_r \sim \mu w_0^2 / (h_r \Delta\gamma) \sim \mu w_0^{20/11} P^{-9/11}. \quad (8)$$

The exponent $-9/11 \approx -0.82$ agrees very well with the powerlaw exponent -0.86 in Fig. 4(a) for the break-up times for temperature-independent material parameters. The temperature dependence of viscosity considerably steepens the dependence of t_r on P . The exponent $20/11 \approx 1.82$ agrees very well with the powerlaw exponent 1.86 in Fig. 4(b). Here, the temperature dependence of viscosity plays only a minor role, because of the weak dependence of $\Delta T_{\max}$ on w_0 for constant laser power.

Figure 4(b) shows that t_r may approach a constant in the limit of narrow laser beams. This is due to the thermocapillary stress scaling approximately as w_0^{-1} , whereas the local capillary pressure gradient scales as w_0^{-3} . Thus, the retarding influence of the capillary pressure compensates the thermocapillary driving force on the scale of the spot size. In this case, the slowly decaying part of the temperature distribution at $r > w_0$ drives the thinning, which depends on P but is independent of w_0 . We refer to this phenomenon as *capillary suppression*.

In summary, we performed a quantitative and systematic experimental and numerical study of the laser-induced rupture of partially wetting liquid films. We obtained excellent agreement using a disjoining pressure potential determined independently. Generally, narrower laser beams and higher powers lead to faster break-up and narrower deformation zones. Dry-spot nucleation in tri(ethylene glycol) films of few micrometer thickness is feasible within less than 10 ms. In immersion lithography, the lower viscosity of water should provide an additional significant speed-up. The effect of the temperature dependence of viscosity is very strong at higher laser powers.

For the considered case of optically thin layers, the film thinning dynamics is insensitive to small variations of the

initial film thickness and the substrate thickness, which makes infrared laser illumination a robust process tool. Moreover, the timescale for reaching the steady state value of the temperature gradient is much shorter than the time-scale for saturation of the maximum surface temperature. This is advantageous for heat-sensitive substrates, as the full force driving the film thinning becomes available quickly, whereas a potentially damaging temperature rise can be avoided by selecting a sufficiently short illumination time.

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