Beyond the thin-film equation: multi-component lattice Boltzmann simulations for film rupture

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(Dated: March 28, 2022)

Under the condition of partial surface wettability, thin liquid films can be destabilized by small perturbations and rupture into droplets. The thin film equation (TFE) is one of the most popular theoretical approaches to describe this mechanism, but it is limited in principle to small equilibrium contact angles and ratios $\epsilon = h/L$ of film height $h$ and lateral size $L$. Here, we use a multi-component lattice Boltzmann (LB) approach to investigate systematically the stability of thin films in contact with a flat surface for large equilibrium contact angles and finite heights. The dispersion law predicted by the TFE turns out to be in very good agreement for angles as large as $130^\circ$. The requirement of a small height is instead a more strict condition for the validity of the TFE, and agreement with LB results is found only for $\epsilon \lesssim 10^{-3}$. The presence of two fluid components allows for a much richer phenomenology than in the single component case, which reflects into the possibility of tuning the dewetting dynamics over a wide range of time scales.

I. INTRODUCTION

Dewetting is the spontaneous, reverse process of wetting of a liquid spreading on a solid surface \cite{1,4}. This phenomenon can be observed in everyday life, for example, when pouring oil on a cooking pan, covering a glass surface with a water film, or when a tear film wets our eyes \cite{5}. Controlling the dewetting dynamics is key to several industrial applications including printable photovoltaics \cite{6,8}, or lubrication and coating processes \cite{9,11}. In general, dewetting dynamics takes place when a thin liquid film, in contact with a partially wettable surface, ruptures into droplets. In the language of thermodynamics, the film reaches its equilibrium droplet shape because the latter is energetically favorable with respect to the flat interface \cite{12,13}.

Dewetting can happen because of intrinsic or extrinsic rupture mechanisms. Extrinsic mechanisms include rupture due to surface heterogeneities or the presence of impurities on the surface \cite{14,15}. These extrinsic mechanisms are opposed to intrinsic, spinodal dewetting ones. Spinodal dewetting occurs in extremely thin liquid films that break up spontaneously due to interface perturbations or thermal fluctuations \cite{10,22}. Several experimental works studied the evolution of thin films on horizontal partially-wettable surfaces \cite{23,24}, chemically structured walls \cite{27}, films with toroidal shape \cite{28} or surrounded by a second viscous phase \cite{29}. Pulsed-laser-induced dewetting of metal alloys has also been investigated \cite{30,31}. The most popular model to describe dewetting phenomena is the celebrated TFE \cite{12}, which describes the space-time evolution of the film height profile $h(x,t)$. In its most simplified form for the deterministic case (i.e., excluding thermal fluctuations) and no-slip boundary conditions, the TFE reads \cite{35,36}:

$$\partial_t h(x,t) = -\frac{1}{3\mu} \mu_x \left[ h(x,t)^3 \mu_x (\Pi(h) + \gamma \nabla^2 h(x,t)) \right],$$ \hspace{1cm} (1)

where $\mu$ is the dynamic viscosity of the film, $\gamma$ its surface tension, and $\Pi(h)$ the disjoining pressure. The disjoining pressure describes the presence of interactions between the fluid and the solid surface, therefore incorporating information on the wetting properties, and is usually expressed in terms of the equilibrium contact angle $\theta_{eq}$. The lubrication approximation \cite{37}, which underpins Eq. (1), assumes the ratio between the characteristic film height $h$ and length $L$ to be very small ($\epsilon = h/L \ll 1$). Furthermore, Eq. (1) is valid under the assumptions of negligible inertial effects (implying a small Reynolds number) and small contact angles due to the inevitable presence, in the theoretical description, of a precursor film. Numerical solutions of the TFE have been obtained using various approaches including contact line solutions \cite{38,40}, gradient dynamics models \cite{41,42}, as well as LB-based methods \cite{22,43}, by allowing to observe the dynamics of $h(x,t)$ within the TFE limits. Other numerical methods have been employed to go beyond the lubrication approximation, including phase-field approaches \cite{45,46}, single-phase LB models \cite{26,47}, and volume-of-fluid methods \cite{49,51}. These approaches take into account inertial effects and control the wettability condition by selecting an equilibrium contact angle rather than introducing a disjoining pressure.

However, a comprehensive investigation of thin-film rupture in a wide range of film height, surface tension and contact angles that can overcome the limitations of the TFE, is still missing. Here, we perform multi-component LB simulations of the thin-film rupture and subsequent dewetting process as sketched in Fig. 1. We systematically explore a wide range of parameters, reaching equilibrium contact angles close to $180^\circ$. As we will show, the
predictions of the TFE in terms of the power spectrum of the film height are in surprisingly good agreement with our simulations for contact angles as high as 130°, and deviations from the TFE prediction are observed from a film thickness $\epsilon$ larger than $3.5 \times 10^{-3}$.

II. METHOD

LB simulations \[52, 53\] solve the Boltzmann transport equation on a lattice. In the long-wavelength limit, Navier-Stokes equations emerge from it as described by the Chapman-Enskog expansion of the discretized Boltzmann equation. We perform simulations of a binary mixture with components labeled A and B. The mesoscopic dynamics of each fluid component $\sigma = A, B$ is described in terms of the probability distribution functions $f^\sigma(x, t)$ of finding a fluid particle in a specific discrete lattice node $x$ and a discrete instant time $t$. The index $i$ refers to a set of discrete velocities $c^i$, which allow the propagation of $f^\sigma$ on the lattice. Here, we employ a D3Q19 lattice, i.e., a set of 19 velocity vectors ($i = 0, \ldots, 18$) on each node of a three-dimensional lattice. Because of the symmetry of the problem, we simulate a quasi-two dimensional system by restricting one dimension of the problem (z) to a lattice of two lattice units. In the LB method, the dynamics of the $f^\sigma_i$ follows the discretized Boltzmann transport equation \[52, 53\]

$$f^\sigma_i(x+c^i,t+1)-f^\sigma_i(x,t)=-\frac{1}{\tau} \left[ f^\sigma_i(x,t)-f^{\sigma,(eq)}_i(x,t) \right]. \tag{2}$$

For the sake of simplicity, in Eq. (2) and hereafter, we fix the lattice spacing $\Delta x$ and the time step $\Delta t$ to one. The left-hand side of Eq. (2) rules the streaming of $f^\sigma_i$ on the lattice, while the right-hand side represents the collision term. This operator models the relaxation of $f^\sigma_i$ towards the discretized local Maxwellian distribution $f^{\sigma,(eq)}_i(x,t)$ with a relaxation time scale $\tau$. The explicit shape of $f^{\sigma,(eq)}_i(x,t)$ is given by \[52\] (repeated indeces are summed up)

$$f^{\sigma,(eq)}_i(x,t) = w_i \rho_\sigma \left[ 1 + \frac{u_{k,e} c^k_i}{c_s^2} + \frac{u_{k,e} u_{j,e} (c^k_i c^j_i - c_s^2 \delta_{kj})}{2c_s^4} \right]. \tag{3}$$

with $c_s = 1/\sqrt{3}$ the speed of sound, and $w_i$ lattice-dependent weights, which, for the D3Q19 lattice, are $w_i = 1/3$ for $i = 0$, $w_i = 1/18$ for $i = 1 \ldots 6$, $w_i = 1/36$ for $i = 7 \ldots 18$, respectively. The fluid component densities $\rho_\sigma$, the total density $\rho$ and the momentum $\rho \mathbf{u}$ can be computed from the populations as $\rho_\sigma = \sum_i f^\sigma_i(x,t)$, $\rho = \sum_\sigma \rho_\sigma$ and $\rho \mathbf{u}(x,t) = \sum_\sigma \mathbf{c}^i f^\sigma_i(x,t)$.

In order to observe phase separation between the two components it is necessary to include fluid-fluid interactions \[52\]. We employ the model proposed by Shan and Chen (SC) \[55, 56\], where a force $F_\sigma(x,t)$ acts on component $\sigma$, entering explicitly in Eq. (2) through a shift in the definition of the momentum:

$$\mathbf{u}_\sigma(x,t) = \mathbf{u}(x,t) + \frac{\tau F_\sigma(x,t)}{\rho_\sigma}. \tag{4}$$

The term $F_\sigma(x,t)$ contains both fluid-fluid as well as wall-fluid interactions. In the SC model, the fluid-fluid interaction term $F^{ff}_\sigma$ takes the form

$$F^{ff}_\sigma(x,t) = -G_{AB} \psi_\sigma(x,t) \sum_i w_i \psi_\sigma'(x+c^i \Delta t, t)e^i, \tag{5}$$

where $\sigma \neq \sigma'$ denote the two components and $G_{AB} > 0$ tunes the (repulsive) interaction strength. Here, the so-called pseudopotential $\psi_\sigma(x,t)$ coincides with the fluid-component density $\rho_\sigma(x,t) = \rho_\sigma(x,t)$. Notice that the implementation of the SC-LB yields a diffuse interface between fluid components with thickness $w_{int}$.

We introduce the wall-fluid interaction following the approach of Huang and coworkers \[57\]. We define the

![Figure 1](image-url)
pseudo-potential for the solid wall as

\[ s(x) = \begin{cases} 
1 & x \in \text{wall} \\
0 & x \in \text{fluid}, 
\end{cases} \tag{6} \]

and, as a consequence, we can write the wall-fluid interactions as

\[ F_{\sigma}^{wf}(x, t) = -G_{w\sigma} \psi_{\sigma} \sum_i w_i s(x + c^i \Delta t) c^i. \tag{7} \]

Eq. 7 can model either a repulsive \((G_{w\sigma} > 0)\) or an attractive \((G_{w\sigma} < 0)\) interaction. To prevent spurious forces generated by the presence of a strong gradient of the components \(A\) and \(B\), we set the value of \(\psi_{\sigma}\) in each wall node to that of the opposite fluid node.

The simulation box is periodic in the \(x\)- and \(z\)-directions, while the fluid is enclosed between two walls along the (vertical) \(y\)-direction. A half-node bounce-back rule implements second-order no-slip boundary conditions at the walls [52]. Hereafter we refer to the liquid film as the \(A\) component, and we report all quantities in lattice units (lbu). We report a validation of the model in Section 1 of the Supplementary Material.

We choose the following set of parameters and initial conditions to study the film rupture: the initial film profile along \(y\) for fluid \(A\) is a step function of width \(h_0\) in the range from 6 to 13 lbu, which drops from \(\rho_A = 1\) (for \(y \leq h_0\)) to \(\rho_A = 0.027\) (for \(y > h_0\)). Conversely, the density of fluid \(B\) raises from \(\rho_B = 0.027\) (for \(y \leq h_0\)) to \(\rho_B = 1\) (for \(y > h_0\)). The interaction parameter \(G_{AB}\) ranges from 1.4 (the minimum value ensuring phase separation) to 1.7, above which the film is always stable and does not rupture. The range of \(G_{AB}\) is directly related to the choice of \(\rho_A\) and \(\rho_B\), as highlighted by the corresponding phase-separation diagram (see Supplementary Material, Fig. S2). In the absence of the wall, these values of \(G_{AB}\) lead to interfacial widths in the range from 2 to 4 lbu (see Supplementary Material, Fig. S2). To ensure the stability of the simulation, we use wall-fluid interaction parameters \(G_{WA}\) and \(G_{WB}\) in the range from \(-0.4\) to \(0.4\). \(G_{AB}\) is directly related to the fluid-fluid surface tension \(\gamma\) [55], which can be measured via Laplace experiments (see Supplementary Material, Fig. S2). The definition of the wall-fluid interactions in Eq. 7 suggests that \(G_{WA}\) and \(G_{WB}\) are related to the wall-fluid interfacial tensions \(\gamma_{WA}\) and \(\gamma_{WB}\), which, however, cannot be measured directly. Instead, for a given value of \(\gamma\), their difference \(\Delta G_W = G_{WB} - G_{WA}\) tunes the value of the equilibrium contact angle \(\theta_{eq}\). The latter is expected to be proportional to \(\Delta G_W\), as observed by Huang and coworkers [52]. Hereafter, all dimensional quantities will be reported in lattice Boltzmann units (lbu). All simulations are performed on a domain of size \(L = 2048\) lbu along the \(x\)-direction and \(H = 256\) lbu along the vertical \(y\)-direction.

![FIG. 2. Stability diagram of thin liquid films in terms of the initial nominal height \(h_0\) and fluid-fluid interaction strength \(G_{AB}\). The corresponding values of \(\epsilon = h_0/L\) and the surface tension \(\gamma\), respectively, are also shown. Three main regions are observed: (1) stability region (pentagons), (2) conditional stability region (triangles), and mixing region (circles). All dimensional quantities are reported in lbu. Further details can be found in the text.](image)

III. RESULTS AND DISCUSSION

We simulate the dewetting dynamics of a thin liquid film placed on a flat wall and the resulting film rupture (see Fig. 1) in the following way: we start with a uniform film of initial, nominal height \(h_0\); we perform a short simulation run to relax this initial condition; due to the diffuse nature of the SC-LB interface, the film height quickly reaches the equilibrium initial (i.e., before the perturbation) height \(h_{in}\), calculated as the distance from the wall at which the density \(\rho_A(y)\) reaches a threshold density \(\rho_{th}\), which is half of its maximum value. The uniformity of the system artificially stabilizes the film, which reaches a (potentially unstable) equilibrium condition. After this relaxation stage, we perturb the film with a random perturbation of its density in its interfacial region with width \(w_{int}\). The perturbed density then reads

\[ \rho'_A(x, t) = \rho_A(x, t)[1 + P\beta] \quad x \in w_{int}, \tag{8} \]

where the perturbation amplitude \(P = 10^{-3}\), and \(\beta\) is a random variable uniformly distributed in \([-1,1]\). In this way, the film height is also perturbed as \(h'(x, t) = h_{in} + \delta h(x, t)\). We explicitly check that \(P\) is small enough and that the results do not depend on its actual value.

By considering all these ingredients together, we are ready to study the stability conditions for thin liquid films as a function of \(G_{AB}\) (i.e., the fluid-fluid surface tension \(\gamma\)), and \(G_{WA}\) and \(G_{WB}\) (i.e., the wetting conditions). We compute stability diagrams by checking the conditions under which the film is stable or ruptures after the initial perturbation.

In Fig. 2 we show the stability diagram as a function of \(h_0\) (and the corresponding ratio \(\epsilon = h_0/L\)) and \(G_{AB}\) (with the corresponding surface tension \(\gamma\)). We can distinguish three main regions: pentagons refer to a stable
film under all wetting conditions, triangles to conditional stability, for which at least one choice of the wetting conditions triggers the film rupture, and circles to cases where the surface tension is not strong enough to preserve the phase separation. In the latter case, the two components (which would demix in the absence of the wall) start mixing.

The cases of conditional stability require to explore further the effect of the wetting conditions. \( G_{WA} \) and \( G_{WB} \) affect the stability in a complex way due to the simultaneous influence of \( h_0 \) and \( G_{AB} \). In Figs. 3 and 4, we show the conditional stability region (and the equilibrium contact angle \( \theta_{eq} \) for the unstable cases) along two cuts. One cut is performed at variable \( h_0 \) and fixed \( G_{AB} = 1.5 \) (i.e., \( \gamma = 0.059 \) lbu) and the other one is performed at variable \( G_{AB} \) and fixed \( h_0 = 6 \) lbu. In all panels of Figs. 3 and 4, the white areas refer to the condition in which the film is stable upon perturbation.

An increase of \( h_0 \) at fixed \( G_{AB} \) and an increase of \( G_{AB} \) at fixed \( h_0 \) both result in an overall improved stability of the film. The overall trend does not come as unexpected. However, while \( G_{WA} \) and \( G_{WB} \) play a symmetric role in

FIG. 3. Contact angle \( \theta_{eq} \) as a function of the wetting conditions \( G_{WA} \) and \( G_{WB} \) corresponding to a vertical cut of Fig. 2 i.e., values are reported at fixed fluid-fluid interaction strength \( G_{AB} = 1.5 \) (\( \gamma = 0.059 \) lbu) and different initial nominal height \( h_0 \) (and \( \epsilon \)). White regions refer to the condition in which the film is stable upon perturbation.

FIG. 4. Contact angle \( \theta_{eq} \) as a function of the wetting conditions \( G_{WA} \) and \( G_{WB} \) corresponding to a horizontal cut of Fig. 2 i.e., values are reported at fixed initial nominal height \( h_0 = 6 \) lbu (\( \epsilon = 2.9 \times 10^{-3} \)) and different coupling parameter \( G_{AB} \) (i.e., surface tension \( \gamma \)). White regions refer to the condition in which the film is stable upon perturbation.
the determination of the contact angle, as already noticed by [57], they play a different role in the stability region. According to the TFE, the film stability should depend only on the equilibrium contact angle \( \theta_{eq} \), that is, on \( \Delta G_w \). [35, 36, 57]. However, the TFE is valid for a single-component fluid only. This result highlights the role played by each of the components on the dewetting dynamics: the best conditions to induce the transition are those for which the wall repels one component (the film), and the other one is attracted by it.

After clarifying these points, we are now in the condition to investigate the dewetting process at large contact angles and check the validity of the TFE beyond its original limits. The time-dependent structure factor of the height profile provides an insightful route to this aim. Starting from the perturbation growth \( \delta h(x, t) = h_{in} - h(x, t) \) and its Fourier transform

\[
\hat{\delta h}(q, t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \delta h(x, t)e^{-iqx} \, dx, \tag{9}
\]

FIG. 5. Power spectra \( S(q, t) \), normalised to the initial one \( S(q, 0) \) for three instants of time (different colours/symbols), different \( h_0 \) and \( \theta_{eq} \). For cases with \( h_0 \leq 7 \text{ lbu} \), a comparison with the TFE predictions is also shown (solid black lines). Vertical dashed lines refer to the fastest \( (q_0) \) and the critical \( (q_c = \sqrt{2h_0}) \) growing modes. All panels have the same q-range.

| \( h_0 \) (lbu) | \( \theta_{eq} \) (°) | \( h_{in} \) (lbu) | \( h_{eff} \) (lbu) | \( q_0 \) (lbu) |
|---|---|---|---|---|
| 6 | 180 | 4.23 | 5.25 | 0.111 |
| 6 | 130 | 2.79 | 4.07 | 0.096 |
| 6 | 90 | 2.69 | 4.57 | 0.073 |
| 7 | 180 | 5.31 | 8.81 | 0.061 |
| 7 | 130 | 3.91 | 5.85 | 0.055 |
| 7 | 90 | 3.77 | 5.16 | 0.050 |
one can define the structure factor (i.e., the power spectrum of the perturbation) as

\[ S(q, t) = |\delta h(q, t)|^2. \tag{10} \]

In Fig. 5 we show \( S(q, t) \) measured for three different heights \( h_0 = 6, 7 \text{ and } 8 \text{ lbu} \) and three different equilibrium contact angles \( \theta_{eq} \geq 90^\circ \), normalized to the initial value \( S(q, 0) \).

The reported values are the result of averages over about 100 independent runs, taken at three different times \( t_1, t_2 \text{ and } t_3 \), with \( t_1 \) corresponding to an early time after the perturbation (but with \( t_2 \) large enough for \( S(q, t_1)/S(q, 0) \) to be appreciably different from 1), \( t_3 \) to a time near the rupture, and \( t_2 \) an intermediate time. These times are different for each panel, and are reported in the Supplementary Material, Tab. S1.

The structure factors \( h_0 = 6 \text{ lbu} \) have the qualitative features expected from the TFE as they show a maximum (defining the fastest growing mode \( q = q_0 \)) followed by a steep descent towards 1 in \( q = q_c \), the so-called critical growth mode. At larger values of \( q \) the normalized structure factor is smaller than one, meaning that the modes at \( q > q_{m} \) decrease in amplitude with time. For values of the contact angle smaller than about 130\(^\circ\), \( S(q, t) \) does not only agree qualitatively with the prediction of the TFE, but also quantitatively. The solid black lines reported in Fig. 3 show the prediction of the deterministic TFE \( S(q, t)/S(q, 0) = e^{\omega(q)t} \), where the dispersion relation \( \omega(q) \) is given by

\[ \omega(q) = \frac{\gamma h_{eff}^4 q_0^4}{3\mu} \left[ 2 \left( \frac{q}{q_0} \right)^2 - \left( \frac{q}{q_0} \right)^4 \right]. \tag{11} \]

Here, the fitting parameter \( h_{eff} \) is an effective initial film height, which does not exactly coincide with the value of \( h_{in} \), but it is close to it (see Tab. I). The only real fitting parameter used here is the fastest growing mode value \( q_0 \), which is not directly modelled within the LB approach. In the deterministic TFE \( q_0 \) is expected to be time-independent. This fact is in agreement with our data for \( h_0 = 6 \text{ and } 7 \text{ lbu} \). The vertical dashed lines in Fig. 5 show the values of \( q_0 \) and the critical growing mode \( q_c \), following the TFE predictions (\( q_c = q_0\sqrt{2} \), with unstable modes being present for \( 0 < q < q_c \)). The match is superior when both \( h_{in} \) and \( \theta_{eq} \) are small (top-right panel of Fig. 5), as the assumptions underlying the TFE are more closely satisfied. Even in these cases, however, there are low-\(q \) tails that are not well reproduced by the TFE. These tails could be the effect of the LB method’s finite (albeit low) compressibility as \( S(q, t) > S(q, 0) \) for the minimum \( q \) implies an enlargement of the overall width of the film. When \( \theta_{eq} = 180^\circ \) the data depart from the TFE prediction that \( q_c = q_0\sqrt{2} \). The agreement achieved for \( h_0 = 6 \) is gradually lost at \( h_0 = 7 \) and it is completely lost at \( h_0 \geq 8 \) (that is, \( \epsilon > 3.5 \times 10^{-3} \)), where more complex behavior emerges that includes even a time dependence of \( q_c \), for \( \theta_{eq} = 180^\circ \).

The dewetting dynamics can be strikingly different even if the equilibrium contact angle is the same for two different wetting conditions. In Fig. 6(a) we report the rupture time \( t_r \) as a function of the wetting conditions for the selected case \( h_0 = 6 \text{ lbu} \) and \( G_{AB} = 1.5 \) (i.e., \( \gamma = 0.059 \text{ lbu} \)). We define \( t_r \) as the time when the film density close to the wall becomes lower than the threshold \( \rho_{th} \), at least at one lattice node. By comparing the left panel of Fig. 4 with Fig. 6(a), we note that wetting conditions driving to the same \( \theta_{eq} \) lead to different rupture times. More in detail, we report in Fig. 6(c) \( t_r \) as a function of one of the two
wall-fluid interaction strengths $G_{WA}$ or $G_{WB}$ for three different cuts of panel (a), namely: a diagonal cut (circles, $G_{WA} = -G_{WB}$); a vertical cut (triangles, $G_{WB} = -0.4$); and a horizontal cut (pentagons, $G_{WA} = 0.4$). For all cases $t_r$, follows a behaviour as a function of the wetting parameter (dotted lines in Fig.3(c)) that we fit with an exponential function to facilitate the reader’s eye. However, the rate can be dramatically different depending on the wetting parameter, even when the equilibrium contact angle is the same, as shown in Fig.3(b) [38]. This result confirms that the dynamics is not simply driven by $\theta_{eq}$, but by a more complex combination of the two wall-fluid interactions. If the film is subject to a strong repulsion ($G_{WA} = 0.4$), the other component has a less prominent effect. On the contrary, if the other component is strongly attracted to the wall ($G_{WB} = -0.4$), a slight variation in the film-wall interaction contributes to a substantial change in the rupture dynamics.

IV. CONCLUSIONS

We performed numerical simulations of the stability and rupture dynamics of liquid films on flat solid surfaces using Shan-Chen multi-component lattice Boltzmann simulations. We characterized the stability conditions of the film in terms of initial film height, surface tension, and equilibrium contact angle. We improved the current understanding of dewetting dynamics spanning a wide range of these parameters up to full dewetting. The theoretical predictions of the thin film equation turn out to be valid up to angles of $130^\circ$, as long as the film thickness $\epsilon = h/L$ is smaller than about $3.5 \times 10^{-3}$. In this sense, the thin film equation requirement of a small contact angle seems necessary only for the description of post-breakup dynamics. As long as the film is intact, wetting conditions corresponding to large equilibrium contact angles are still well described by the thin film equation. Our analysis of the stability and rupture times underlines the richness of phenomenology brought in by the presence of two liquid components. The presence of two components introduces a much richer phenomenology and opens the possibility of tuning the dynamics of dewetting over a wide range of time scales by controlling the wetting parameters of the two fluids separately.

ACKNOWLEDGMENTS

This work has received financial support from the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) – Project-ID 431791331 – SFB 1452. Furthermore, the authors acknowledge financial support by the DFG within the priority program SPP2171 “Dynamic Wetting of Flexible, Adaptive, and Switchable Substrates”, projects HA-4382/11 and SE-3019/1-1. The authors gratefully acknowledge the Gauss Centre for Supercomputing e.V. (www.gauss-centre.eu) for funding this project by providing computing time on the GCS Supercomputer JUWELS at Jülich Supercomputing Centre (JSC).

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