On the Dewetting of Liquefied Metal Nanostructures

Shahriar Afkhami and Lou Kondic
Department of Mathematical Sciences, New Jersey Institute of Technology, Newark, NJ 07102 USA

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Abstract. Direct numerical simulations of liquefied metal nanostructures dewetting a substrate are carried out. Full three-dimensional Navier-Stokes equations are solved and a volume-of-fluid method is used for tracking and locating the interface. Substrate wettability is varied to study the influence of the solid/liquid interaction. The effects of initial geometry on the retraction dynamics is numerically investigated. It is shown that the dewetting velocity increases with increasing the contact angle, and that the retraction dynamics is governed by an elaborate interplay of initial geometry, inertial and capillary forces, and the dewetting phenomena. Numerical results are presented of the dewetting of nanoscale Cu and Au liquefied structures on a substrate.

Keywords: Liquefied Metal Nanostructures, Dewetting, Navier-Stokes, Surface Tension, Volume-of-Fluid

1. Introduction

Modeling of fluid problems characterized by length scale measured in 10s to 100s of nanometers involves considering a number of issues, in particular when free boundaries are involved. This modeling is further complicated in the setups involving the presence of fluid/solid interfaces. One member of a family of problems where these effects are relevant involves evolution of liquefied metals. Very recently, these materials have been extensively considered for the synthesis and assembly on nanoscale (see e.g. (Habenicht et al., 2005; Habenicht et al., 2008; Roberts et al., 2013)). The dewetting of liquefied gold (Au) nanoparticles are experimentally considered by Habenicht et al. (2005; 2008), for example. In these physical experiments, the long time dynamics of the nanodroplet is governed by the competition of the inertial, capillary, and surface forces, giving rise to droplet contraction toward a global energy minimizer; if the inertial forces overcome the adhesion energy and dissipative losses, the nanodroplet is ejected from the substrate on a time scale of the order of 10 ns. The small length and short time scales involved in this process make the experimental study of the dewetting dynamics a daunting task.

Discrete molecular dynamics (MD) approaches can be utilized to simulate the systems on nanoscale (Fuentes-Cabrera et al., 2011a; Fuentes-
Cabrera et al., 2011b). Fuentes-Cabrera et al., for example, carried out MD simulation using a Lennard-Jones (LJ) interface potential to study various wetting regimes of nanoscale liquefied copper (Cu) nanodisks on graphite (Fuentes-Cabrera et al., 2011a). They showed that the dewetting velocities are consistent with inertial flow estimates, and that thicker and smaller radius nanodisks have proportionately lower velocities. They have also found that for a particular choice of the LJ potential, the dewetting nanoparticles jump off from the graphitic substrate with velocities that are similar to those observed experimentally in (Habenicht et al., 2005) for Au liquefied nanostructures on glass.

While the MD approach is very powerful, it is computationally expensive. An alternative approach is to use continuum modeling although it is unclear that the assumptions of continuum fluid mechanics are satisfied on these very short spatial and temporal scales. In addition to concerns related to validity of continuum models on nanoscale, modeling of fluid structures on substrates involves additional degree of complexity since one needs to carefully include the liquid/solid interaction forces, which may be dominant on such short length scales. Time-dependent evolution necessarily involves consideration of contact lines, where liquid, solid, and gas phase meet and modeling of the physics in the vicinity of contact lines is demanding and challenging, due to well known issues related to the so-called contact line singularity (de Gennes, 1985; Haley and Miksis, 1991). Due to all these complexities, typically the problem is considered within the long-wave theory, although in the context of liquid metal films this approach is questionable due to large contact angles involved. Despite the concerns about its validity, this approach has turned out to be very useful, providing significant insight to the dynamics and stability properties of fluids on nanoscale, including liquefied metals (Trice et al., 2007; Ajaev and Willis, 2003; Kondic et al., 2009).

An important property of the numerical schemes that needs to be developed for NS equations on nanoscale is the ability to accurately and robustly deal with the fluid-fluid as well as fluid-solid interactions. Furthermore, capillary forces are often very important on nanoscale, and therefore the efficacy of a numerical scheme is closely dependent on the ability of both the surface tension discretization and the flow solver in minimizing the introduction and amplification of errors. Accurate computation of the geometrical properties of the interface such as interfacial normals and curvature, sharp surface force representation, and accurate and stable simulation are key ingredients in feasible computation on nanoscale. Here, we use the ‘volume-of-fluid’ (VoF) interface capturing methodology, which is a widespread approach for representing fluid interfaces and, when coupled to a flow solver, is also used to compute.
quantities related to surface tension that enter the flow calculation. Until recently, however, the VoF method was not generally deemed appropriate for the study of phenomena in which surface tension is the driving force (e.g. the wetting and dewetting of solid surfaces, as occurs in droplet and spray technologies as well as in various natural phenomena). However, recent improvements to calculating curvature and applying the surface tension force appear to resolve this issue (Afkhami and Bussmann, 2008; Afkhami and Bussmann, 2009; Afkhami et al., 2009).

In this paper, we use numerical simulations based on full three-dimensional (3D) NS equations based on a VoF method to study the dewetting of liquefied nano-sized structures on substrates with varying wettability. The motion of the fluid both inside the drop and in the surrounding flow are numerically modeled. Unlike most of other theoretical and computational studies, the Reynolds number and the contact angle need not be small. In this work, we explore whether our continuum formulation will result in simulations consistent with MD simulations using LJ potentials. We initially focus our simulations on the dewetting behavior of liquid Cu nanodisks and ring-shaped nanostructures. We will then consider Au triangular nanostructures, that do not possess radial symmetry.

2. Mathematical Model

The equations of conservation of mass, \( \nabla \cdot \mathbf{u} = 0 \), and momentum, \( D(\rho \mathbf{u})/Dt = \nabla \cdot (-p \mathbf{I} + \mathbf{\tau}) + \mathbf{F}_{st} \) (NS equations), govern the fluid motion in the drop and the surrounding gas. Here \( \mathbf{u} \) is the velocity vector, \( p \) is the pressure, \( \rho \) is the fluid density, \( \mathbf{\tau} \) is the shear stress tensor, and \( \mathbf{F}_{st} \) is the surface tension force (per unit volume). The shear stress tensor is defined as \( \mathbf{\tau} = \mu (\nabla \mathbf{u} + (\nabla \mathbf{u})^T) \), where \( \mu \) represents the fluid dynamic viscosity. The flow equations are written in an Eulerian frame of reference, and thus a solution of these equations must be coupled with a methodology for following the deforming fluid-fluid interface. Here, the VoF volume tracking algorithm is implemented (Hirt and Nichols, 1981; Gueyffier et al., 1999; Afkhami and Bussmann, 2009). Volume tracking requires the introduction of a scalar function \( f \), defined as \( f = 1 \) inside the drop and \( f = 0 \) outside the drop, for a two fluid system. Since \( f \) is passively advected with the flow, it satisfies the advection equation, \( Df/Dt = 0 \). Density and viscosity are then evaluated via volume-weighted formulae as \( \rho = \rho_g + (\rho_d - \rho_g)f \) and \( \mu = \mu_g + (\mu_d - \mu_g)f \), respectively, where subscripts \( d \) and \( g \) refer to the drop and gas phases, respectively. Surface tension is reformulated as an equivalent
Figure 1. Adaptive mesh: the mesh is adaptively refined around the contact line (solid line) and areas of high capillary pressure. Color contours depict the pressure distribution colored according to the reference pressure in the gas phase, with a maximum value colored in dark red a minimum value colored in dark blue.

volume force as originally proposed in the ‘continuum surface force’ (CSF) model (Brackbill et al., 1992) as, \( \mathbf{F}_{st} = \sigma \kappa \delta_{S} \mathbf{n} \), where \( \sigma \) is a constant interfacial tension and \( \delta_{S} \) denotes the Dirac delta function for the surface separating the fluids. The curvature \( \kappa \) and unit normal \( \mathbf{n} \) are geometric characteristics of the surface and are described in terms of \( f \) and computed with a second-order ‘height-function’ (HF) method (Sussman, 2003; Cummins et al., 2005; Afkhami and Bussmann, 2008; Francois et al., 2006). If partial slip is allowed at the contact line, we will impose the Navier-slip boundary condition at the (static) solid surface \( y = 0 \), \( (u, w)|_{y=0} = \lambda \partial(u, w)/\partial y|_{y=0} \), where \( \lambda \) is the slip length.

To investigate the effects of the relevant dimensionless parameter, we nondimensionalize the governing equations using a velocity scale \( V = \sqrt{\sigma/\rho L} \), where \( L \) is the appropriate length scale. The resulting time scale, \( T = \sqrt{\rho L^3/\sigma} \), is proportional to the time it takes for the capillary wave to propagate across the drop. With these choice of scales, the NS equations now can be written in nondimensional form \( D(\rho u)/Dt = \nabla \cdot (-\rho \mathbf{u} + \text{Oh}^{-1} \mathbf{u}) + \mathbf{F}_{st} \). \( \text{Oh} = \eta/\sqrt{\rho \sigma L} \) is the Ohnesorge number, representing the ratio of the time it takes for momentum to diffuse across the droplet to the period of the capillary wave.

3. Numerical Model

A complete description of the numerical methodologies can be found in (Popinet, 2003; Afkhami and Bussmann, 2008; Afkhami and Bussmann, 2009). Our in-house code is built based on an early version of the open-source flow solver GERRIS (see (Popinet, 2003)). Briefly, the domain is
discretized adaptively using the oct-tree finite volumes arranged hier-
archically (see Fig. 1). Primitive variables are collocated at cell centers;
normal velocities are also specified at faces, as in the MAC grid. The
flow equations are discretized using a projection method based on a
fractional-step scheme. In the fractional-step projection method, an
interim velocity is computed first; this velocity field is projected onto
a divergence-free velocity field, with the pressure field obtained as the
solution of a Poisson equation. Advection terms are discretized using
a second-order upwind scheme (Bell et al., 1989). As the face-centered
velocities are exactly divergence-free, the volume fractions are advected
using these velocities. The numerical algorithm for the imposition of
the contact angle is described in details in (Afkhami and Bussmann,
2009). In this algorithm, the value of the contact angle affects the
overall flow calculation in two ways: it defines the orientation of the VoF
reconstruction in cells that contain the contact line and it influences
the calculation of $F_{sl}$ by affecting the curvature calculated in cells at
and near the contact line to balance the surface tension forces on the
contact line as in Young’s relation (Young, 1805): $\sigma \cos \theta_{eq} = \sigma_{sg} - \sigma_{sl}$
where $\sigma_{sg}$ and $\sigma_{sl}$ are interfacial tensions for the solid-gas and solid-
liquid surfaces, respectively, and $\theta_{eq}$ is the equilibrium contact angle.
If the actual contact angle is out of equilibrium, the resulting mis-
match in curvature generates a force thus driving the contact line to a
configuration that satisfies Young’s relation.

3.1. Computational Setup

We consider three geometrical configurations. Figure 2 shows the initial
configurations for the case of a Cu disk (a), a Cu ring (b), and a Au
triangular (c) nanostructures. We denote the disk geometry with an
initial height $h_0$, and radius $R_0$; the ring geometry with an initial outer
radius $R_{out}$ and inner radius $R_{in}$; the triangle geometry with an initial
long side $a$, short side $b$, and thickness $h_0$; the initial contact angle is
denoted as $\theta_0$. The computational domain is $L_x \times L_y \times L_z$ and the
maximum grid resolution of the adaptive mesh is denoted by \( \Delta \). For all the simulations, an open boundary condition (pressure and velocity gradient equal zero) is imposed at the top and a symmetry boundary condition is imposed on the lateral sides. For nanodisk and nanoring liquid structures, a free-slip boundary condition is specified at the substrate in accordance with the MD simulations in (Fuentes-Cabrera et al., 2011a). For triangular nanostructures, the Navier-slip boundary condition with \( \lambda = 3 \) nm is imposed, so that the computed detachment velocities are consistent with the experimental results (Afkhami and Kondic, 2013; Habenicht et al., 2005).

Table I. Overview of the sets of parameters used in the numerical simulations (Fuentes-Cabrera et al., 2011b; Habenicht et al., 2005).

<table>
<thead>
<tr>
<th>Material</th>
<th>( \rho (\text{kg m}^{-3}) )</th>
<th>( \eta (\text{kg m}^{-1} \text{s}^{-1}) )</th>
<th>( \sigma (\text{kg s}^{-2}) )</th>
<th>( \ell_c (\text{nm}) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Air</td>
<td>1.22</td>
<td>1.98 \times 10^{-5}</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cu</td>
<td>7900</td>
<td>4.28 \times 10^{-3} (\text{at 1500 K})</td>
<td>1.30</td>
<td>1.78</td>
</tr>
<tr>
<td>Au</td>
<td>17310</td>
<td>4.25 \times 10^{-3} (\text{at 1500 K})</td>
<td>1.15</td>
<td>0.90</td>
</tr>
</tbody>
</table>

4. Results

4.1. Dewetting of Cu Nanostructures

4.1.1. Nanodisks

We first consider a Cu disk of \( h_0 = 15 \) Å, \( R_0 = 150 \) Å, and \( \theta_0 = 90^\circ \), when the equilibrium contact angle \( \theta_{eq} = 80^\circ \) is imposed. The nanodisk dewets the substrate and collapses into a spherical cap with an equilibrium base radius \( R_{eq} = 8.65 \) nm corresponding to a contact angle of \( 80^\circ \). We set \( L_x = L_y = L_z = 375 \) Å. Numerical simulations are carried out on an adaptive mesh (see Fig. 1) with a maximum grid resolution of \( \Delta = 2.93 \) Å. We compute the retraction of the disk and consequent coalescence into a nanodroplet. We track the position of the edge of the retracting rim \( R(t) \) as a function of time. Figure 3 shows the results of direct simulations (–). We also plot the simulation results with \( \Delta = 1.46 \) Å (–) to show convergence with mesh refinement. After 140 ps (not shown in Fig. 3), the Cu nanodrop reaches an equilibrium state with \( R_{eq} = 8.63 \) nm. The final stage is followed by the contact line oscillations about the equilibrium state; a signature of inertially-dominated convection. To gain additional insight regarding the effects that drive
oscillations, we have carried out additional set of simulations with modified inertial effects. This was implemented by varying incompressible fluid density, but more generally the dynamics can be characterized by Ohnesorge number, here defined as \( Oh = \eta/\sqrt{\rho \sigma R_0} \). For the Cu nanodisk, \( Oh \approx 0.35 \), suggesting that inertial effects are important; the same conclusion can be reached by considering an intrinsic length scale, \( \ell_v = \eta^2/(\rho \sigma) \), above which inertial effects become significant; for liquid Cu, \( \ell_v \approx 1.78 \text{ nm} \), therefore smaller than typical length scales considered here. As an example, we also present the computational results for \( Oh \approx 1.1 \) (--) in Fig. 3. As shown, reduced inertial effects eliminate the final stage oscillations.

Figure 4(a-c) shows sequences of snapshots of the simulations at various times for \( \theta_{eq} = 80^\circ, 115^\circ, \) and \( 140^\circ \). For \( \theta_{eq} = 80^\circ \), viscous forces overcome the capillary force during dewetting until the drop assumes its equilibrium shape. For \( \theta_{eq} = 115^\circ \), the nanodisk quickly dewets the surface and at about 40 ps the retracting rim has already collapsed into a droplet that begins to elongate in the \( y \)-direction. The collapsing nanodrop then reaches to a maximum elongation at around 100 ps. After that, the droplet rebounds and settles at an equilibrium state with \( \theta_{eq} = 115^\circ \) at around 250 ps. We also notice that the contraction in the \( x \)-(horizontal)-direction and the elongation in the \( y \)-direction are now more pronounced compared to what was found for \( \theta_{eq} = 80^\circ \). A different type of evolution occurs when \( \theta_{eq} \) is increased beyond \( 130^\circ \).
Figure 4. Snapshots of the Cu nanodisk with $h_0 = 15$ Å, $R_0 = 150$ Å, and $\theta_0 = 90^\circ$. Direct numerical simulations of NS equations for $\theta_{eq} = 80^\circ$ (a), $115^\circ$ (b), and $140^\circ$ (c). MD simulations (d) employing a Lennard-Jones potential corresponding to $\theta_{eq} = 138.5^\circ$, from (Fuentes-Cabrera et al., 2011a).

For $\theta_{eq} = 140^\circ$, the contraction is so fast that the nanodrop jumps off the surface, following elongation in the $y$-direction. This process begins with the accumulation of the liquid material towards the center of nanodroplet, leading to a vertical movement of the center of mass. The nanodroplet detaches from the substrate at about 100 ps. Figure 4(d) shows the results of MD simulations in (Fuentes-Cabrera et al., 2011a), when employing a Lennard-Jones potential that corresponds to $\theta_{eq} = 138.5^\circ$. We find quantitative agreement with our continuum simulations.
Figure 5. $x$-$y$ cross sections of a Cu disk ($h_0 = 15$ Å, $R_0 = 150$ Å, $\theta_{eq} = 140^\circ$) at $t = 0, 5, 10, 20, 30, 40, 110$ ps with (a) $Oh \approx 0.35$ and (b) $Oh \approx 1.1$. The case (a) results in a drop that eventually detaches from the substrate as in Fig. 4(c), while for (b) the final outcome is a sessile droplet.

Figure 5(a-b) further illustrates the relevance of inertial effects. Here, we compare the $x$-$y$ cross sections for the case $\theta_{eq} = 140^\circ$ when $Oh \approx 0.35$ and 1.1. As shown, reduced inertial effects eliminate the ejection of the nanodrop from the surface. For $Oh \approx 0.35$ (Fig. 5(a)), a capillary ridge is formed at the initial stage and propagates from the nanodisk rim along the drop-gas interface progressing to the top of the droplet, causing a rapid rise in the droplet height and eventual ejection, while for $Oh \approx 1.1$ (Fig. 5(b)), the capillary ridge is less pronounced, leading to a weaker collapse and no ejection.

We also study the effect of the equilibrium contact angle, $\theta_{eq}$, on the retraction time and the upward velocity. Table II presents the variation of the retraction time and the upward velocity with $\theta_{eq}$. As shown, the retraction dynamics is slower for smaller $\theta_{eq}$, and the upward velocity increases with increasing $\theta_{eq}$. These values are fully consistent with the values obtained in (Fuentes-Cabrera et al., 2011a) using MD simulations.

It is also instructive to compare the velocity field for various $\theta_{eq}$. Figure 6 shows the velocity field at 20 ps for $\theta_{eq}=140^\circ$ and at 40 ps for $\theta_{eq}=80^\circ$ where the position of the edge of the rim is the same in both cases. As shown, the magnitude of the velocities are larger for $\theta_{eq}=140^\circ$ than of those for $\theta_{eq}=80^\circ$. In this figure, the background depicts the
Table II. Effect of contact angle on the retraction time and the upward velocity.

<table>
<thead>
<tr>
<th>$\theta_{eq}$</th>
<th>130°</th>
<th>140°</th>
<th>150°</th>
</tr>
</thead>
<tbody>
<tr>
<td>collapse time (ps)</td>
<td>120</td>
<td>100</td>
<td>90</td>
</tr>
<tr>
<td>upward velocity (m s$^{-1}$)</td>
<td>62</td>
<td>119</td>
<td>137</td>
</tr>
</tbody>
</table>

Figure 6. Velocity vectors of a Cu nanodisk dewetting ($h_0 = 15$ Å, $R_0 = 150$ Å, $\theta_0 = 90°$) (a) at 20 ps for $\theta_{eq}=80°$ and (b) at 40 ps for $\theta_{eq}=140°$; the front has moved about the same distance in both cases. Color contours show the pressure field distribution colored according to the reference pressure in the gas phase, with a maximum value colored in dark red a minimum value colored in dark blue.

4.1.2. Nanorings
We next consider the collapsing of Cu liquefied nanorings. When a ring of liquid is placed on a substrate, it retracts and collapses into a static droplet due to capillary effects (González et al., 2013). We show here the direct computational results for the dewetting of Cu liquefied nanorings as $\theta_{eq}$ is varied. We consider the ring geometry with an initial outer radius $R_{out} = 150$ nm, inner radius $R_{in} = 75$ nm, and $\theta_0 = 60°$ (see Fig. 2). The radii and the $\theta_0$ are chosen so that the drop volume remains almost unchanged from the previous nanodisk.
Figure 7. Early-time retraction velocities of a Cu nanodisk dewetting ($h_0 = 15 \text{ Å}$, $R_0 = 150 \text{ Å}$, $\theta_0 = 90^\circ$): $\theta_{eq} =$ 80° ($\square$), 115° ($\triangle$), 130° ($\bullet$), and 140° ($\diamond$).

case. The nanoring structure is out of equilibrium and the capillary pressures resulted from the inner and outer curvature imbalance drives an inward flow which eventually leads to a collapse of the ring into a static drop. We note that here $Oh \approx 0.4$ calculated based on the average radius, i.e. $(R_{\text{out}} + R_{\text{in}})/2 = 75 \text{ nm}$. We set $L_x = L_y = L_z = 375 \text{ Å}$. Numerical simulations are carried out on an adaptive mesh (see Fig. 1) with a maximum grid resolution of $\Delta = 2.93 \text{ Å}$.

Figure 8 shows the time evolution of the nanorings for $\theta_{eq} =$ 45°, 60°, 75°, and 90°. In particular, we are interested in the time, $\tau$, it takes for a ring to collapse, defined as the time when the inner radius goes to zero. Figure 9 shows $\tau$ as a function of $\theta_{eq}$. We find that a larger $\theta_{eq}$ leads to a smaller collapse time. We also observe that $\tau$ scales approximately linearly with $\theta_{eq}$.

4.2. Dewetting of Au Nanostructures

4.2.1. Nanotriangles

Recently, Habenicht et al. (Habenicht et al., 2005; Habenicht et al., 2008) experimentally examined whether it would be possible to begin with initially deformed droplets, in the form of solid flat triangular gold structures, on the surface and observe dewetting induced detachment. They considered Au triangular structures on the SiO$_2$ substrate. When melted by laser irradiation, the energetically unfavorable initial structure coalesces into a droplet. The transformation of surface energy to
Figure 8. Snapshots of the Cu nanoring with $R_{\text{out}} = 150$ Å, $R_{\text{in}} = 75$ Å, and $\theta_0 = 60^\circ$, for $\theta_{eq} = 45^\circ$ (a), $60^\circ$ (b), $75^\circ$ (c), and $90^\circ$ (d). The frames on very right show the drop at the time at which the collapse takes place, with inner radius going to zero. $Oh \approx 0.4$.

kinetic energy drives the motion and under appropriate conditions, detachment of the resulting droplets was observed. Figure 10 (Fig. 1(c) in (Habenicht et al., 2005)) shows sequences of images of the development of the dewetting process from flat triangles toward spherical structures.

Here, we simulate the experiments of Habenicht et al. (2005). The initial triangle geometry has equal sides $a = 405$ nm and the initial contact angle $\theta_0 = 90^\circ$ with $\theta_{eq} = 140^\circ$ (equilibrium contact angle of liquid Au on SiO$_2$). The computational domain is $L_x = L_y = L_z = 494$ nm and the maximum grid $\Delta = 1.5$ nm. At the substrate, the Navier-slip with slip length of 3 nm is imposed; this particular value is obtained
Figure 9. The effect of $\theta_{eq}$ on the collapse time, $\tau$, for a Cu nanoring on the substrate ($R_{out} = 150 \, \text{Å}$, $R_{in} = 75 \, \text{Å}$, and $\theta_0 = 60^\circ$). $Oh \approx 0.4$.

Figure 10. Fig. 1(c) in (Habenicht et al., 2005). Initially solid triangular gold structure with a thickness of 50 nm and a side length of 800 nm dewets on the substrate and coalesces into a droplet. $\theta_{eq} = 131^\circ$.

by direct comparison with the experiments of Habenicht et al. (2005). Figure 11 represents the droplet velocity as a function of the initial thickness $h_0$. Our numerical results reproduce very well the experimental findings in (Habenicht et al., 2005). In particular, we note that both experiments and simulations show decreasing ejection velocity as the initial thickness is increased. This trend can be intuitively understood by realizing that the upward momentum is gained by the transformation of surface to kinetic energy. Thus, increasing the initial thickness results in a lower upward velocity by decreasing the surface to volume ratio. We observe that both the simulations and the experimental results show approximately linear dependence of the ejection velocity on $h_0$, with the simulation predicting zero ejection velocity for $h_0 \geq 200 \, \text{nm}$.

Figure 12 shows sequences of simulations of the dewetting of initially triangular nanostructures when initial thickness is varied. The consequent detachment of the droplet at various times is also demonstrated. As shown, the dewetting process begins at the corners, where
the curvature is high. Due to a high surface tension force and large equilibrium contact angle, the liquid starts to accumulate at the corners. Vertices accumulate more mass than the straights sides. The humps at the corners then coalesce into a droplet. Finally, owing to a low viscosity of liquid gold ($\ell_v \approx 0.9$ nm), inertia dominates over viscous dissipation giving rise to an upward movement that leads to droplets detaching from the surface with velocities of tens of meters per second (see Fig. 11). As shown, increasing the initial thickness will result in a slower dynamics and an increase in the collapsing time. The time scale of the retraction process is consistent with the experiments of (Habenicht et al., 2005) measuring a detachment time scale of the order of 10 ns.

Figure 13 shows the pressure distribution at the (substrate) $x$-$z$ plane for (a) $h_0 = 24$ nm and (b) 47 nm. Solid line represents the line of contact and the background demonstrates the adaptive grid used. The high surface tension force due to high curvature increases the capillary pressure at the corners. This pressure gradient results in a transversal velocity driving the traveling rims. The time of which all the rims coalesce depends on the pressure gradient, explaining the difference of collapsing times shown in Fig. 12. As also shown, the sides of the triangle reach the center first, followed by vertices. This asynchronicity is the reason for excitation of contact line oscillations during the dewetting process, as illustrated in Fig. 13.

Finally, we explore the effect of the initial shape. We consider an isosceles nanotriangle geometry with an initial long side $a = 488$ nm, short side $b = 247$ nm, $h_0 = 24$ nm, $\theta_0 = 90^\circ$, and $\theta_{eq} = 145^\circ$. Figure
Figure 12. Snapshots of the Au equilateral nanotriangles with $a = 405$ nm, $\theta_0 = 90^\circ$, and $\theta_{eq} = 140^\circ$, for $h_0 =$ 24 nm (a), 47 nm (b), 95 nm (c), and 142 nm (d). The nanotriangular structure collapses into a droplet and detaches from the substrate (see Fig. 11 for corresponding ejection velocities).

14(a) shows the sequences of the numerical simulation. As shown, the dewetting behavior is different compared to dewetting dynamics of an equilateral triangle, although both processes lead to the ejection of the formed droplet from the substrate. Figure 14(b) shows the pressure distribution at $x$-$z$ plane. It shows a higher capillary pressure at the vertex of the long edges due to a higher curvature. However, the triangle small edge vertices arrive faster at the center than the vertex of the longer sides. This mismatch excites the oscillation of the droplet translating into a tumbling movement after the droplet leaves the substrate.
5. Conclusion

We show that the direct numerical simulation based on Navier-Stokes equations can be utilized to study dewetting of liquefied metal nanostructures. In particular, we find that early-time dynamics of the dewetting of a Cu nanodisk is inertially dominated, and that viscous dissipation cannot be expected to be the dominant resistance to the short-time dewetting. Furthermore, we demonstrate that the equilibrium contact angle, $\theta_{eq}$, plays a key role in the dewetting dynamics. For sufficiently large $\theta_{eq}$, the retraction and the consequent collapse into a nanodroplet is followed by the detachment of a nanodrop from the substrate with an upward velocity that is an increasing function of $\theta_{eq}$. Considering Cu nanorings, we show that $\theta_{eq}$ influences the retraction velocity and the collapsing time. Our numerical results based on a VoF formulation is consistent with the results of the MD simulations offering a potential venue for modeling dewetting phenomena on nanoscale. We also directly simulate the dewetting process of initially triangular Au nanostructures that coalesce into nanodroplets which in turn detach from the surface. We demonstrate that the dynamics of the dewetting behavior is consistent with the experimental observations and that the computed detachment velocity and time are also in agreement with experimental measurements. We expect our findings to be of direct relevance to nanomaterial processes involving rapid dewetting.

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Figure 13. Contours of the pressure field distribution and the adaptive mesh at x-z plane for the Au equilateral nanotriangles with $a = 405$ nm, $\theta_0 = 90^\circ$, and $\theta_{eq} = 140^\circ$. (a) $h_0 = 24$ nm and (b) 47 nm. The high capillary pressure at the corners are shown. The pressure distribution is colored according to the reference pressure in the gas phase, with a maximum value colored in dark red a minimum value colored in dark blue.
Figure 14. (a) Snapshots of the Au isosceles nanotriangle structure with $a = 488$ nm, $b = 247$ nm, $h_0 = 24$ nm, $\theta_0 = 90^\circ$, and $\theta_{eq} = 145^\circ$. The nanotriangular structure collapses into a droplet and detaches from the substrate at around 5 ns. (b) Contours of the pressure field distribution and the adaptive mesh at $x$-$z$ plane. The pressure distribution is colored according to the reference pressure in the gas phase, with a maximum value colored in dark red a minimum value colored in dark blue.