Oscillating friction of nanoscale capillary bridge

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Abstract: The presence of a capillary bridge between solid surfaces is ubiquitous under ambient conditions. Usually, it leads to a continuous decrease of friction as a function of bridge height. Here, using molecular dynamics we show that for a capillary bridge with a small radius confined between two hydrophilic elastic solid surfaces, the friction oscillates greatly when decreasing the bridge height. The underlying mechanism is revealed to be a periodic ordered-disordered transition at the liquid–solid interfaces. This transition is caused by the balance between the surface tension of the liquid–vapor interface and the elasticity of the surface. This balance introduces a critical size below which the friction oscillates. Based on the mechanism revealed, a parameter-free analytical model for the oscillating friction was derived and found to be in excellent agreement with the simulation results. Our results describe an interesting frictional phenomenon at the nanoscale, which is most prominent for layered materials.

Keywords: capillary bridge; nano-confinement; elasticity; atomically smooth surface

1 Introduction

A capillary bridge, or liquid bridge, is always present between two solid surfaces in contact with or near each other under ambient conditions [1, 2]. Its presence can be due to either thermally activated capillary condensation which is in thermodynamic equilibrium with vapor [3–5], or other non-spontaneous ways, e.g., a tree frog climbing trees by excreting mucus to form capillary bridges between its feet and the tree bark. The great significance of these capillary bridges is not only due to their universal presence, but also their key contribution to the forces between surfaces [6]. The effect of capillary bridges on adhesion has been widely studied since the first observation of a capillary bridge [7]. At the nanoscale, their presence causes interesting behavior which is different with that at the microscale. For example, Cheng and

Robbins found that due to molecular layering of the liquid confined in the narrow gap between the spherical tip and substrate, the adhesion force oscillates strongly with respect to the height of the bridge [8, 9]. Later Valenzuela et al. show that the oscillation can be suppressed by tuning the values of practical parameters, e.g., tip elastic constant and approach speed [10].

Besides its effect on the adhesion force, the capillary bridge also strongly affects friction between two solid surfaces [11, 12]. In micro- or nano-electromechanical systems (M/NEMS), the capillary bridge often prevents devices from functioning under ambient conditions or leads to damage in their fabrication processes due to the large friction it induces [13]. For example, when the height of the capillary bridge is less than twice the typical atomic sizes, e.g., 0.5 nm, it increases the friction greatly as the water confined between

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the surfaces becomes ice-like [14–17]. However, under certain circumstances, the capillary bridge may serve as a lubricant, reducing the intrinsically large friction between solid surfaces [18]. Sometimes, the presence of a capillary bridge can even lead to counter-intuitive friction phenomena. For example, at low sliding velocity, the capillary-mediated friction decreases logarithmically with velocity and increases with temperature [19, 20]. The mechanism for such intriguing phenomena was revealed to be a combination of thermally activated condensation and the rupture process of the capillary bridge [21, 22].

At present, most of the studies to understand the effects of capillary bridges on friction focus on systems with either multiple capillary bridges or an individual macroscopic capillary bridge. There are limited studies at the nanoscale. By using noncontact dynamic force spectroscopy, Lee et al. studied the shear interaction of a single capillary bridge with its height varying from 6 to 25 nm and found that the shear interaction can be divided into elastic and damping forces due to contact angle hysteresis [23]. With atomic force microscopy or surface force apparatus (SFA), the friction of a capillary bridge with a diameter in micrometers has been experimentally studied and showed that the friction varies continuously with the height of the bridge as expected by macroscopic/continuum theory [24, 25]. There is little study on the friction caused by a capillary bridge with a height comparable to the size of single atom and a diameter in nanometers. Such a capillary bridge will be present when two smooth hydrophilic surfaces [26] come into contact. Another case where capillary bridges could play key roles is the capillary channel, such as a carbon nanotube filled with discrete liquid columns [27].

We systematically studied the effect of the capillary bridge on friction at the nanoscale. Using molecular dynamics (MD) simulation, we found that for a capillary bridge with a small radius confined between two hydrophilic elastic solid surfaces, the friction oscillates dramatically when decreasing the bridge height. The underlying mechanism was revealed to be a periodic ordered-disordered transition at the liquid–solid interfaces caused by the balance between the capillary force of the bridge and the

bending stiffness of the surface. A parameter-free model was proposed based on such understanding and found to be in good agreement with the MD simulation results.

2 Methods

As shown in Figs. 1(a) and 1(b), the system is composed of two smooth solid surfaces with a nonvolatile capillary bridge confined between them. The surfaces are created by separating the (001) crystal planes of a face-centered-cubic (FCC) crystal cuboid where $[1\ 0\ 0]$, $[0\ 1\ 0]$, and $[0\ 0\ 1]$ direction is along the x , y , and z axis, respectively. Each surface is composed of two layers of atoms. For a typical simulated system, there are 400 atoms for the capillary bridge and 3,200 atoms for the solid surfaces, i.e., 800 atoms within each solid layer. The mass of an individual atom in the solid surface is set to that of carbon atom. Since there are 1,600 atoms per plate, the mass per plate is 3.2×10^{-23} kg.

The bottom layer of the lower surface is fixed to the stationary stage. The top plate (slider) is coupled to a driven tip through three independent springs connecting the center of mass of the top plate with the tip. The force exerted on the top plate by the springs is evenly distributed among all the atoms within the top plate. The corresponding spring constants are $k_x = k_y = 7$ N/m and $k_z = 28$ N/m, which are chosen to stand for the weak mechanical interactions with the substrate [26, 28]. To avoid vibration of the upper plate, viscous damping η is applied to the three springs with values corresponding to critical damping. The springs together with the damping provide reasonable constraints for the dynamics of the plate. A periodic boundary condition is applied in both the x and y direction. A vacuum along the z direction is used to prevent interaction between the system and its images.

The interaction between atoms in the surfaces is described by a harmonic potential $U_{\text{har}} = k(r - d)^2 / 2$, where k is the spring constant and r is the distance between neighboring atoms. The equilibrium bond distance $d = a / \sqrt{2}$ with a being the lattice constant of the conventional unit cell of the FCC structure. The bond springs are connected between the atoms

and their nearest neighbors. For the liquid atoms, we first describe their interactions with the Lennard–Jones (LJ) potential as $U_{LL} = 4\epsilon[(\sigma/r)^{12} - (\sigma/r)^6]$, where ϵ is the depth of the potential well and σ is the distance at which the inter-particle potential is zero. In addition, to eliminate evaporation, we further combine every four liquid atoms into a short molecule chain. Such a constraint is achieved by applying a finitely extensible nonlinear elastic (FENE) potential $U_{FENE} = -1/2KR_0^2 \ln[1 - (r/R_0)^2]$ on the neighboring atoms within one single chain [8], where K is the bead spring coefficient and R_0 is the maximum extent of the bond. As verified by Cheng and Robbins [9], the vapor pressure of the liquid in this case can be neglected without significantly increasing the molecular size or slowing down the dynamics. Aiming at capturing the generic behavior of the system, the mass of the liquid atoms is chosen to be the same as that of the solid. The interaction between the solid and liquid is described by the LJ potential with $U_{SL} = 4\epsilon_{SL}[(\sigma_{SL}/r)^{12} - (\sigma_{SL}/r)^6]$, where $\epsilon_{SL} = 0.8\epsilon$ and $\sigma_{SL} = 1.2\sigma$. For both U_{LL} and U_{SL} , the LJ potential is truncated and shifted at a cutoff radius $r_c = 2.5\sigma$.

As we are interested in the properties of the system at steady state, we first equilibrate the system at T for 20 million timesteps with the height of the tip remaining fixed. During this process, the distance between the two surfaces, h , is calculated as the vertical distance between the center of mass of the two inner layers (labeled red in Fig. 1). Then the

tip was moved at a constant velocity, v_{dr} , along the $+y$ direction with the height of the tip remaining fixed. When the temperature and momentum of the capillary bridge were stable, another 100 million timesteps were run for statistics. All simulations were performed using the large-scale atomic-molecular massively parallel simulator (LAMMPS) developed at Sandia National Laboratories [29].

In our simulations, we used dimensionless quantities with their units described in terms of ϵ , σ , and m . With $\epsilon = 43.7$ meV, $\sigma = 0.5$ nm, and $m = 1 \times 10^{-25}$ kg, the typical values of the quantities used are $a = 2^{-1/3}\sigma$, $v_{dr} = 0.04\sigma/\tau \approx 10$ m/s where $\tau = \sqrt{m\sigma^2/\epsilon} \approx 1.88$ ps, $k = 228\epsilon/\sigma^2$, $K = 30\epsilon/\sigma^2$, $R_0 = 1.5\sigma$, and $T = 350$ K. The corresponding time step of the velocity Verlet algorithm is 0.002τ which is about 4 fs.

3 Results and discussion

Figure 1(c) shows the lateral force, F_L , i.e., friction, and normal force F_N exerted on the sliding surface by the capillary bridge as a function of the distance between surfaces, h . The volume of the nonvolatile confined liquid is kept constant for systems with different h . Both F_L and F_N show damped oscillations as h increases and are out of phase with each other. For F_N , its periodicity is due to molecular layering of the liquid confined in the narrow gap between the tip and substrate [8]. For F_L , we note that similar behavior has been observed for two surfaces with

Fig. 1 (a) A schematic of the simulation system; the liquid is embedded between two solid plates. The upper plate is pulled by a spring with the other moving at constant speed, and the lower plate is fixed. (b) Each plate is composed of two layers of atoms. The outer layers (grey) were frozen and the inner layers (red) were equilibrated at room temperature. (c) The magnitude of shear and normal force experienced by the plate as a function of the distance between plates.

the inter-surface space filled with liquid [30]. In this case, there is no capillary bridge and the oscillation is attributed to the ratio between the lattice size of the solid surface and the diameter of the liquid molecules [30].

To verify whether the mechanism as revealed in Fig. 1(c) also holds for our observations, we first calculated the number density of the liquid. As shown in Fig. 2(a), a layered structure is observed within the liquid. As h decreases from 2.69 to 1.77, the number of layers gradually decreases from 4 to 3, indicating a complete structural transition period happens in between. We calculated the structure factor for the layer of liquid next to the solid surface $S(q)$. The structure factor is calculated as $S(q) = \frac{1}{N} \sum_{j=1}^N \sum_{k=1}^N e^{iq(r_j - r_k)}$, where r_j and r_k represent the position of the j^{th} and k^{th} atom, N is the amount of liquid atoms within the first layer, q is the Hash lattice vector of the solid, and $\langle \rangle$ represents the average value for the system in equilibrium [31, 32].

Clearly, within this layer of liquid, the structure changes periodically as h decreases (Figs. 2(b)–2(d)), showing that this is the reason for the oscillating behavior in F_L . The atomic structure corresponding to this transition is shown in Fig. S1 in the Electronic Supplementary Material (ESM). At the comensurate state, the capillary bridge experiences large friction, showing a stick-slip phenomenon, whereas at the disordered state, the force trace becomes erratic (Figs. 2(e) and 2(f)). The transition between ordered and disordered states accounts for the periodic changes in F_L . Also, as h decreases, the contact area between the capillary bridge and the solid surface increases, resulting in an inversely proportional relationship between F_L and h . Together with the ordered-disordered transition, these two mechanisms lead to the damped oscillating dependence of F_L on h as shown in Fig. 1(c). In the systems studied here, due to the ultra-smooth surface, the friction originates solely within the contact area between the liquid and solid. This indicates a different

Fig. 2 (a) In a typical periodic phase for the height varying from 2.69 to 1.77 ($h=2.69, 2.45, 2.31, 2.18, 2.09, 2.00, 1.88, 1.77$), the number of liquid layers changes from 4 to 3; this number can be easily obtained by counting the peaks. Correspondingly, the structure factor between the solid and the neighboring liquid layer is shown in (b–d), which indicates an ordered- (b for 4 layers, d for 3 layers) disordered (c for 3 layers) transition. (e, f) Lateral force and normal force for $h=2.45$ and 1.77 , respectively.

mechanism from the stick-slip friction phenomenon observed when sliding an etched gold tip across a mica surface, which is caused by the three-phase contact line [23].

Compared with a slit filled with liquid, one distinct feature of the capillary bridge is its lateral size characterized by the contact radius r , i.e., the radius of its contact area with the solid surface. To investigate the dependence of F_L and F_N on r , we increase r with h being fixed. In this case, the volume of the capillary bridge is increased. Interestingly, we find that there is a critical r_c above which the dependence of F_L on h becomes continuous (Fig. 3) while for F_N the discontinuity still holds. The remaining of discontinuity in F_N is easy to understand as it is decided by the instability of the system when h increases. For liquid confined between surfaces, obviously this critical behavior is unique to the capillary bridge, and to our best knowledge, this critical radius phenomenon has never been reported.

The key to understanding the existence of the critical radius is the capillary force present near the contact line [33] along with the Laplace pressure, which is unique to capillary bridges. The capillary force induces a normal force on the surface, $F_\gamma = 2\pi\gamma r \sin\theta$, where γ is the surface tension of the liquid–vapor interface and θ is the contact angle. The Laplace pressure also exerts a normal force on the surfaces, $F_{\Delta P} = \pi r^2 \Delta P$, where ΔP is the pressure drop across the surface of the capillary bridge. From the Young–Laplace theory, the pressure drop can be estimated as $\Delta P = 2\gamma\kappa = \gamma(1/r_1 + 1/r_2)$, where r_1 and r_2 are the principle curvatures of the surface of the capillary bridge. Thus, the force generated by the Laplace pressure is $F_{\Delta P} = \pi r^2 \gamma (1/r_1 + 1/r_2) =$

$\pi\gamma r(r/r_1 + r/r_2)$. For hydrophilic surfaces as studied here, it is trivial to show that $r_1 = r - h/2 \cdot (1 - \tan\theta/2)/(1 + \tan\theta/2)$ and $r_2 = h/2\cos\theta$. By noting that the critical radius r_c is much larger than the distance between surfaces h , i.e., $r \gg h$ when $r = r_c$, $r_2 \leq h/2 \ll r$, we have $r/r_1 + r/r_2 \gg 2$. In this case $F_{\Delta P} \gg 2\pi\gamma r \geq F_\gamma$. Given that the total force exerted on the surfaces by the capillary bridge $F_{\text{cap}} = F_\gamma + F_{\Delta P}$ and $F_{\Delta P} \gg F_\gamma$, F_{cap} can be approximated as

$$F_{\text{cap}} \approx F_{\Delta P} = \pi\gamma r(r/r_1 + r/r_2) \approx 2\cos\theta\pi\gamma r^2/h = \pi r^2\Delta P \quad (1)$$

by noting that $r_1 \approx r$. The approximated pressure drop $\Delta P = 2\gamma\cos\theta/h$ used in Eq. (1) has been adopted by de Gennes [7].

Evidently, the force F_{cap} induces deformation of the surfaces as shown in Fig. 4(a) and illustrated in Section 2 of the ESM. According to the Kirchhoff–Love plate theory $\nabla^2\nabla^2 w = \Delta P/D$, where w is the deflection of the surface and D is the bending stiffness. Bending stiffness is determined by $D = EH^3/12(1-\nu^2)$, where E is the elastic modulus, H is the thickness of the plate, and ν is Poisson ratio. According to the solution given by Poisson in 1829 [34], $w = \Delta P(x^2 - r^2)^2/64D$, where x is the distance to the center of the contact area. When $x = 0$, we get the maximum deflection $w_m = 3(1-\nu^2)\gamma\cos\theta r^4/8EH^3h$.

When w_m is large enough, within the contact area between the solid surface and the liquid, different regions experience different lateral forces as shown in Fig. 4(b); the overall resistance exerted on the sliding surface will be the average of all the lateral forces. For example, in our case when $w_m \geq \delta_0 \approx \delta/5$, all states corresponding to different lateral

Fig. 3 Comparison of F_L and F_N for a capillary bridge with (a) $r=9.52$ and (b) $r=11.50$ when h is 5.0. As r increases, the oscillating dependence of F_L on h disappears, but remains for F_N .

Fig. 4 (a) Schematic of the deformation of the plate and the capillary bridge under the pressure differential caused by surface tension. The magnitude of the deformation is enlarged for better visibility. (b) The corresponding shear force as a function of height.

forces are covered (corresponding to points with $2.43 < h < 2.76$ in Fig. 4(b)). In this case, the discontinuity in F_L is eliminated. Thus, the critical condition for the elimination of the discontinuity in F_L is $3(1-\nu^2)\gamma\cos\theta r^4/8EH^3h=w_0$, where w_0 is the minimum range within which the lateral force reaches its maximum and minimum as h changes for a capillary bridge with a small radius and height (in our case $w_0=\delta/5$). Thus, the critical radius predicted theoretically, r_c , can be estimated as

$$r_c = (8EH^3hw_0/3(1-\nu^2)\gamma\cos\theta)^{1/4} \quad (2)$$

In the system studied, $E=240$, $\nu=0.33$, $H=1.62$, $\delta=0.78$, $\gamma=0.88$, $\theta=75^\circ$, $h=5.0$, and $w_0=\delta/5$; the critical radius predicted theoretically is $r_c=10.12$. This is in excellent agreement with the results from the direct MD simulations which were $9.52 < r_c < 11.50$. It should be noted that the transition only holds when F_L shows a discontinuous dependence on h for a capillary bridge with small radius, i.e., h is small (comparable to σ).

As shown in Eq. (2), our model predicts that the critical condition is also determined by the elastic modulus and the contact angle. In the MD simulation, the elastic modulus can be adjusted by changing the harmonic potential parameter K , and the contact angle can be adjusted by changing the depth of the potential well of the LJ potential ϵ_{LS} between the solid and liquid. By changing E and θ within the direct MD simulations, we plotted the phase diagram for the transition in F_L as a function of h (Fig. 5). The critical transition estimated from the MD simulations are in excellent agreement with the prediction by our analytic model (Eq. (2)).

In experiments with h on the order of nanometers,

Fig. 5 Phase diagram of the shear modes characterized by contact angle and elastic modulus. The stepped mode is in red and the continuous mode is in blue. The critical transition as predicted by Eq. (2) is plotted as a solid curve.

the radius of capillary bridge is usually larger than the critical radius, which makes it hard to observe the oscillation in the lateral force. However, with our understanding as quantitatively summarized in Eq. (2), it is possible to observe the oscillation by controlling the radius of the capillary bridge, the distance between the surfaces, the elastic modulus of the surface, and by using a certain liquid. As a rule of thumb for experimentalists, in Table 1, we list the critical radius, r_c , for some typical friction pairs with confined liquid at the interface [35]. A potential experimental system could be graphene layers deposited by exfoliation onto a smooth surface (e.g., a Si/SiO₂ surface), which represent a large category of experimental systems [26, 36, 37]. For such systems, it has been shown experimentally that the few-layer graphene is partly freely suspended between hills of the substrate [26]. Thus, when two hydrophilic surfaces [38] of this kind slide against

Table 1 Critical radius r_c for some typical capillary bridge systems, H is about 1 nm in each condition, $E^*=E/(1-\nu^2)$, the effective elastic modulus.

Solid	Liquid	E^* (GPa)	θ (°)	δ (nm)	γ (nN/nm)	h (nm)	r_c (nm)
Graphene	Water	~2,000	85	0.3	0.073	0.3–1.6	11.1–16.8
Silicon	Water	~200	40	0.3	0.073	0.3–1.6	3.6–5.5
Gold	Water	~100	60	0.3	0.073	0.3–1.6	3.4–5.1
Glass	[C4C1im][NTf2]	10	25	~1	0.04	0.1–0.5	2.0–3.0
Silicon	[C4C1im][NTf2]	~200	35	~1	0.04	0.1–0.5	4.2–6.3

each other under ambient conditions, their frictional process may be well described by our model.

4 Conclusions

In recent years, there have been some studies about the properties of liquid bridges. However, there are very limited studies on the friction of capillary bridges at the nanoscale [23–25, 27, 39]. For a capillary bridge between hydrophilic surfaces, we observed an oscillating dependence of friction on the distance between surfaces using MD simulations. The behavior of the normal force in our simulation coincided with the findings of Cheng and Robbins [9]. Moreover, with a large scale simulation, we verified that the friction varies continuously with the height of the bridge as expected by macroscopic continuum theory [24, 25]. Interestingly, there is a critical size below which the friction oscillates. Usually, macroscopic capillary theory [40] and elasticity theory [41] can be extended to the nanoscale. However, we found an entirely different change of friction at the nanoscale compared to the macroscale.

The underlying mechanism was revealed to be the balance between the capillary force of the capillary bridge and the bending stiffness of the surface. The parameter-free analytical model proposed agrees well with the simulation results, thus providing a guideline for controlling friction between hydrophilic surfaces at distances on the order of nanometers, e.g., dip-pen nanolithography where smooth kinetics is required for the tip movement [42].

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