Dewetting Properties of Metallic Liquid Film on Nanopillared Graphene

Xiongying Li, Yezeng He, Yong Wang, Jichen Dong & Hui Li

Key Laboratory for Liquid-Solid Structural Evolution and Processing of Materials, Ministry of Education, Shandong University, Jinan 250061, P. R. China.

In this work, we report simulation evidence that the graphene surface decorated by carbon nanotube pillars shows strong dewettability, which can give it great advantages in dewetting and detaching metallic nanodroplets on the surfaces. Molecular dynamics (MD) simulations show that the ultrathin liquid film first contracts then detaches from the graphene on a time scale of several nanoseconds, as a result of the inertial effect. The detaching velocity is in the order of 10 m/s for the droplet with radii smaller than 50 nm. Moreover, the contracting and detaching behaviors of the liquid film can be effectively controlled by tuning the geometric parameters of the liquid film or pillar. In addition, the temperature effects on the dewetting and detaching of the metallic liquid film are also discussed. Our results show that one can exploit and effectively control the dewetting properties of metallic nanodroplets by decorating the surfaces with nanotube pillars.

Wettability is a fundamental property of a solid surface, and plays important roles in daily life, industry and agriculture. The amazing climbing ability of geckos1–3, as well as the self-cleaning effects of lotus leaves and butterfly wings4,5, are all related to the unique micro- and nanostructures on their surfaces. Inspired by these strategies, functional surfaces with special wettability, such as nanopillared surfaces, have elicited great research interest6–9 and been synthesized by various methods10–14. For example, the superhydrophilic surface15,16 with a water contact angle of almost 0° fabricated by UV irradiation has been successfully used as a transparent coating with antifogging and self-cleaning properties. On the other hand, various phenomena, such as contamination, snow sticking, erosion and water transport are expected to be inhibited on superhydrophobic surfaces17–19 with a contact angle larger than 150°.

Much research has been performed to investigate the wetting behavior of liquid nanostructures, including water, polymers and metals20–28. In terms of metallic thin films, these materials have attracted much attention not only due to their central importance for thin film technology, such as controlling the homogeneity of thin films and coatings29,30, but also because of their potential applications in catalysis, photovoltaics, sensor, transparent conductors and memory cells31–33. Due to the dramatic difference in the physical properties relative to typical polymer and water films, including high surface energy, low viscosity, and differences in the long range interaction forces, continuous metallic films contract far from equilibrium, and are unstable with respect to forming discrete nanodroplets via dewetting34. Numerous experimental and theoretical investigations have confirmed that the dewetting begins at the edges or corners of films where the surface tension is high35–37, and the dewetting evolution undergoes three successive stages: rupture of the film resulting in the occurrence of nanoholes, growth of the holes followed by the mass and energy transmissions, and finally the decay of the holes. According to hydromechanics, inertia plays a dominant role in the dewetting dynamics of metallic nanostructures36,37. Recently, a unique property of these films, i.e. jumping off the substrates, has been reported. Habenicht et al.36 first demonstrated that deformed Au droplets could convert their surface deformation energy into kinetic energy, which makes them contract and finally detach from the non-wettable substrate. Fuentes-Cabrera et al.38, Afkhami and Kondic37 have reported that the evolution of the liquid Cu film is strongly dependent on its initial shape and the contact angle (CA). Such a jumping phenomenon had also been found in the evaporation of water droplets on G39. These important findings shed new light on the interfacial interactions and possibly pave the way for self-assembly nanotechnology40–42.

Much research has been done for the interfacial behavior of graphene, such as wetting and applications of super-hydrophobicity in self-cleaning materials, however, to our knowledge, fewer efforts have focused on the dewetting-induced detachment of nanodroplets, especially how to use super-hydrophobicity to control the movement of nanodroplets. Wetting and dewetting are important practical issues in physical and material sciences, and provide an opportunity for comprehensive and satisfactory understanding of the interfacial...
properties of nanomaterials. As of yet, although there has been much progress in the understanding of dewetting, some important questions concerning the super-hydrophobic surfaces remain unanswered. Motivated by this and inspired by the amazing wettability of the nanofabricated surfaces mentioned above, we performed molecular dynamics (MD) simulations to explore the dewetting behavior of metallic thin films on pillared graphene (PG) consisting of uniform-distributed carbon nanotube (CNT) pillars, with the aim of controlling the contraction and detachment of liquid films by tuning the sizes of nanopillars. Figure 1 shows the model of the simulated system that a liquid Cu disk is deposited on a nanopillared graphene (PG) constructed with vertically aligned carbon nanotube (CNT) pillars.

Results
To determine the role of the capped CNT nanopillars on the dewetting properties of surfaces, a comparison between G and PG surfaces is made. Figure 2 shows snapshots of the dewetting dynamics and potential energy distributions of liquid Cu disks with \( D = 135.6 \) Å. Notably, when \( H = 3 \) Å, the film on either the G or PG first ruptures to form some nanoholes (Figure 2a and b). These nanoholes gradually combine into a large hole, then decay and disappear. This phenomenon provides insight into the evolution process of nanoholes within the ultrathin liquid film (see the video 1 in the Supporting Information for details of the Cu-PG system), which is in good agreement with the experimental results. When \( H \) increases to 5 Å, no holes can be observed. This indicates that whether the nanohole exists during the contraction is mainly dependent on the \( H \) of the film rather than the surface structure. Interestingly, further research presents that the PG can detach the nanodroplet from it (see the video 2 in the Supporting Information), while this detachment cannot be observed on G, regardless of the fact that \( H \) increases to 10 Å. Like PG, another substrate PG2 displays similar dewetting properties, as shown in Figure 2c. The droplets detach from the PG and PG2 surfaces at 41 and 51 ps, respectively. Importantly, the film sizes (initial diameter \( D \), thickness \( h \) and radius of droplet \( R \) used in this study and the detaching time \( t_d \) = 41 ps, are far smaller than those in previous studies. The comparison indicates that the nanopillars would improve the dewetting property of flat surfaces which brings great advantages in detaching droplets from the substrate. These findings are critical in the chemical system, daily life and nanotechnology, as well as guiding nanoparticles toward a tumor. In addition, the switch of dewetting modes with respect to the \( H \) also gives us the inspiration to avoid the formation of nanoholes within the superthin films by decreasing the film sizes, which is very important in the practical applications of superthin liquid films, such as homogeneous thin film, coating, anti-adsorption and self-cleaning materials.

Potential energy distributions indicate that the dewetting begins at the edges of films and nanoholes. As the simulation proceeds, the edge propagates and contracts toward the center of the disk, which makes mass and energy flow and finally forms a spherical droplet when the system approaches the equilibrium (\( t = 200 \) ps). A better understanding of the contraction and detachment of the liquid film can be gained by the energy profiles. The total potential energy, \( E_{\text{tot}} \), is defined as

\[
E_{\text{tot}} = E_{\text{C-C}} + E_{\text{Cu-Cu}} + E_{\text{C-Cu}}
\]

where \( E_{\text{C-C}} \), \( E_{\text{Cu-Cu}} \) and \( E_{\text{C-Cu}} \) represent the C-C, Cu-Cu and C-Cu interaction, respectively. Naturally, the \( E_{\text{C-C}} \) is equal to 0 eV due to the fix of the substrate. The \( E_{\text{Cu-Cu}} \) is a resisting force which hinders the slipping and ejection of droplets due to its reverse direction to the slipping and detachment. Therefore, \( E_{\text{Cu-Cu}} \) is the driving force. The energy difference, \( \Delta E_{\text{Cu-Cu}} \), is partially transformed into kinetic energy of the droplet, which induces contraction and even detachment. Figure 3 gives the \( \Delta E_{\text{Cu-Cu}} \) and \( v_z(t) \) as a function of time. As for those systems without detachment, the \( \Delta E_{\text{Cu-Cu}} \) of the Cu-PG system is much larger (lower) than the Cu-G system, indicating that the PG exhibits a stronger dewettability than the flat G, as shown in Figures 3a and b. At the first 15 ps, the rapid decrease of \( \Delta E_{\text{Cu-Cu}} \) and \( E_{\text{C-Cu}} \) accelerates the contraction of the liquid film and helps the droplet move upward, thus leading to the rapid nearly-linear increase of \( v_z \). It is worth noting that there exists a small peak when \( H = 3 \) Å

![Figure 2](image-url)  
**Figure 2** | Dewetting dynamics and potential energy distributions of Cu films (\( D = 135.6 \) Å) on substrates. (a) G. (b) PG, \( a = 10.2 \) and \( h = 13.67 \) Å. (c) Another pillared graphene (PG2) with the concave-cap CNT as the pillar, \( a = 3.4 \) and \( h = 10.92 \) Å. For clear observation of the internal energy distribution, one forth of some liquid droplets has been cut off. When \( H \) increases to 5 Å, the droplet detaches from the PG surface at 41 ps due to dewetting. Further increasing \( H \) to 7 Å, detachment occurs in the Cu-PG2 system at 51 ps.
(Figure 3c), which is considered to be directly related to the existence of nanoholes, and this plays a critical role in determining the dewetting mode of metallic films. When the liquid film evolves, the inertial force predominates, which is one of the determining factors for the detachment. The high AE_{Cu-Cu} and high equilibrium AE_{Cu-Cu} are the key factors for the detachment. The high AE_{Cu-Cu} could generate a large vz, the low AE_{Cu-Cu} ensures that the vz remains at a constant value above zero.

There still exists the question of what plays the dominant role in determining the contraction and detachment: the inertial force, or the viscous force. To gain insight into this question, we carry out additional analysis from the perspective of hydromechanics. The Reynolds number (Re) is a dimensionless quantity which gives a measure of the ratio of inertial forces to viscous forces, and consequently quantifies the relative importance of these two types of forces during the liquid film evolution. Re is given as Re = \frac{\rho v L}{\mu}, where \rho, v and \mu are the density, mean velocity and dynamic viscosity of the fluid, respectively, and L is a characteristic linear dimension. For liquid Cu in these systems, Re \approx 6.10, indicating that the inertial force plays an important role in the contraction and detachment, which accords well with the deductions from Refs. 36 and 37.

Figure 4 provides the dependency of H on the detachment. Notably, as H increases from 5 to 10 Å, t_d increases monotonously, but v_d slightly decreases with some fluctuations. At every point of H, the t_d (v_d) decreases (increases) significantly with the increasing a. These phenomena indicate that the dewetting dynamics of the Cu liquid film is more sensitive to the a than H. That is because increasing every 1 Å of the H results in a much larger increase of the E_{Cu-Cu} than the E_{Cu-Cu}; however, increasing a conversely leads to the decrease of the hindering force E_{Cu-Cu}. That is, decreasing the E_{Cu-Cu} is the key to effectively detach the liquid droplet. Therefore, we can conclude from Figure 4 that it is an effective means to control the dewetting dynamics of the liquid film by tailing the interval distance of pillars rather than tuning the initial H of the films. The above analysis indicates the great effect of a on detachment. Actually, the height h of pillars may also affect the dewettability. To verify this notion, a series of simulations are carried out on the liquid evolution of Cu films on PGs with h ranging from 13.67 to 17.22 Å. Different from the a, the h presents slight dependency on the detachment. Figure 5a and b shows the influence of a and h on the t_d and v_d. Clearly, the t_d exhibits a nearly exponential increase with the increasing a. However, when a is fixed, both the t_d and v_d change slightly with the h increasing from 13.67 to 17.22 Å. It indicates that the detachment properties of the droplet is not sensitive to the height of the pillars, which may bring interesting insights to the design of novel microfluidic devices.
To explore how the shape of the liquid film affects the dewetting behavior, a comparison among the Cu films in the shapes of disk, square and triangle is made. Figure 6 shows snapshots of these films, as well as the corresponding $t_d$ and $v_d$. From the top view of the dewetting process, it can be seen that the Cu disk always remains circular in shape, and the square Cu forms a circle at about 23 ps, but more than 23 ps for the triangle Cu. Interestingly, the snapshot of the triangle Cu clearly indicates that the contraction at the straight lines is faster than that at the corners. Importantly, the $t_d$ (62 ps) for the triangle Cu is largest, followed by 58 ps for the square, then by 49 ps for the disk. The comparison between these films strongly suggests that the sharp corner does not favor the contraction and detachment.

To investigate the influence of temperature ($T$) on the $t_d$ and $v_d$, we have carried out a series of MD simulations of a Cu disk with $D = 135.6$ Å on the PG at temperatures ranging from 1500 to 2000 K. As shown in Figures 7 a and b, increasing $T$ from 1500 to 2000 K leads to a 24.44% decrement of $t_d$ and 82.76% increment of $v_d$ for $a = 10.2$ Å, indicating that the detachment can be controlled by adjusting the temperature. These temperature effects are attributed to the enhanced inertial effects, which result from the decreases of density and viscosity due to the increase of $T$. As an example, when $T$ increases from 1500 to 1900 K, the $Re$ increases from 6.10 to 10.13, indicating that the role of inertial effects is very remarkable at high temperatures.

Figure 5 | The size effects of nanopillars on detachment: $t_d$ (a) and $v_d$ (b). The solid lines are fit curves, which reveal that the $t_d$ ($v_d$) exponentially increases (decreases) with the interval distance of pillars $a$.

Figure 6 | The initial shape effects of Cu films on detachment. The $H$ is fixed at 10 Å. (a)–(c) Snapshots of the detaching process. (d) The $v_d(t)$ of the liquid Cu droplet when $D = 162.72$ Å. (e) and (f) $t_d$ and $v_d$ as functions of $D$. The black, red and blue lines stand for the results of the disk, square and triangle, respectively.

Figure 7 | The temperature effects on detachment. (a) and (b) The curves correspond to those systems with $H = 10$ Å, $h = 13.67$ Å, and $a$ ranging from 6.8 to 10.2 Å. The dashed lines are fit curves, which reveal that the $t_d$ ($v_d$) presents an exponential relation with $T$. (c) and (d) The effect of the surface structure change on the detaching properties at high temperatures. The sample used here corresponds to the system $a = 11.9$ Å, $h = 13.67$ Å and $H = 5.0$ Å. Results show that the $t_d$ decreases 4.36% and the $v_d$ increases 8.12% after considering the surface structure change, indicating that it is relatively easy for the Cu droplet to jump off with the consideration of the structure change.
surfaces, the HCNT could also detach the droplet, as shown in horizontally placed carbon nanotubes (HCNT). Like the PG and PG2 more sensitive to the films and the substrates. Figure 8 a and b show that the detachment on the PG2 surface displays strong dependence on both
detachment on the PG surface, the 

Figure 8 presents the detaching properties of the Cu liquid droplet on the PG2 surfaces. Like the detachment on the PG surface, the detachment on the PG2 surface displays strong dependence on both the films and the substrates. Figure 8 a and b show that the $t_d$ significantly increases but the $v_d$ slightly decreases with the increasing $H$. Additionally, both the $t_d$ and $v_d$ display an exponential relation with $a$ and $T$.

Figure 9 shows dewetting properties of liquid films on the horizontally placed carbon nanotubes (HCNT). Like the PG and PG2 surfaces, the HCNT could also detach the droplet, as shown in Figure 9a. Further research presents that both the $t_d$ and $v_d$ display an increasing tendency with increasing $D$, as shown in Figure 9b, which is consistent with the results in Figure 6, which reflects the importance of surface nanostructures on the special dewettability and indicates the effective method to control these properties by tuning the sizes of the Cu films.

**Methods**

The Cu nanodisk is extracted from a bulk liquid, then deposited at a distance of 3.0 Å above the PG surface. The CNT pillars are placed on the G to mimic the lotus leaves which possess superhydrophobicity and self-cleaning effects.

MD simulations are performed to study the dewetting properties of metallic liquid films on PG under the constant-volume and constant-temperature (NVT) ensemble, by using the large-scale atomic/molecular massively parallel simulator (LAMMPS) package.

The interaction among metal atoms is described by an embedded atom method (EAM) potential.

The C-C interaction is modeled by an adaptive intermolecular reactive empirical bond order (AIREBO) potential, due to the fact that metal and carbon can form soft bonds via charge transfer from the π electrons in the sp$_2$ hybridized carbon to the empty 4 s states of metal.

we utilize the 12-6 Lennard-Jones (LJ) potential with a well depth $\varepsilon = 0.018$ eV and size parameter $\sigma = 3.0$ Å to describe the metal-C interaction.

This LJ potential was found to reproduce the equilibrium CA as 132° for Cu on pristine graphene. These values are in good agreement with the experimental one 140°.

The temperature is controlled by the Nose–Hoover method and the time integration of the Newton’s equation of motion is undertaken using the velocity Verlet algorithm with a time step of 1.0 fs. All of the substrates are fixed during the MD simulations.

The CA of a liquid metal droplet on surfaces is calculated by the method used by Werder.

Firstly, the isochore line of the time-averaged metal number density being about one fourth of the bulk density is defined as liquid-air interface. Secondly, we fit a circle to the interface, and then measure the CA. The velocity of the liquid nanodroplet along the z direction vertical to the substrate is defined as $v_z(t) = \frac{dZ_{CM}}{dt}$, where $Z_{CM}$ is the displacement of the nanodroplet center of mass, and $t$ is time. The moment when the C-metal potential energy $E_{C-metal}$ equals zero is defined as the detaching time $t_d$. After detachment, the liquid droplets remain at a constant velocity, which is defined as the detaching velocity $v_d$.

**Discussion**

In summary, this biomimetic study on the wettability of nanopillared graphene reveals the importance of the surface nanostructures on the special dewettability. The MD results show that the CNT pillars act as the effective detwetting components, and this not only leads to the increase of dewettability but also brings great convenience in detaching metallic droplets from the surfaces. The inertia plays a dominant role in the dewetting dynamics, especially at high temperature. The detachment can be controlled by simply tuning the geometrical parameters of the films and the surface nanostructures, as well as the temperature. The switching dewetting mode and the controlled detachment not only shed new light on the special dewettability of nanopillared surfaces, but also pave the way for new and promising applications of these surfaces in daily life, self-cleaning materials, nanofluidic devices and homogeneous coatings.

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