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Distinctive nanofriction of graphene coated copper foil

Weihui Wang [a], Qing Peng [b,c], Yiquan Dai [a], Zhengfang Qian [c], Sheng Liu [c,*]

[a] School of Mechanical Science and Engineering, Huazhong University of Science & Technology, Wuhan, Hubei 430074, China
[b] Department of Mechanical, Aerospace and Nuclear Engineering, Rensselaer Polytechnic Institute, Troy, NY 12180, USA
[c] School of Power and Mechanical Engineering, Wuhan University, Wuhan, Hubei 430072, China

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ABSTRACT

We investigate the nanoscale frictional behaviors and characteristics of mono-, bi-, and tri-layer graphene/Cu (1 1 1) systems using molecular dynamics simulations. The existence of graphene layers significantly enhances the load carrying capacity, extends the low friction range, and effectively decreases the friction. The friction coefficient depends on the normal force, which is very distinctive in contrast to classic friction laws and shows the nanoscale effect. The stable friction coefficients are 0.24, 0.18, and 0.11 for monolayer, bilayer, and trilayer graphene, respectively. Such distinguishable nanofriction behaviors suggest an experimental routine to characterize the number of graphene layers on copper foil or other substrates through nanoscratching as an alternative approach to Raman spectroscopy.

1. Introduction

Since its first exfoliation from graphite crystals in 2004 [1], graphene, with its distinctive band structure and fascinating combination of extraordinary properties, has attracted enormous attention in scientific and industrial communities because of its potential for various applications in graphene-based composites [2], nano-devices [3], energy storage materials [4], and so on. As the strongest ever measured [5], gas-impermeable [6], chemically and thermally stable [7,8], and atomically thick material [1], graphene is an excellent candidate as a protective coating or solid lubricant for enhancing surfaces, reducing adhesion, friction, and wear when coated on various surfaces so as to extend the longevity of graphene-based devices [9–13].

The microscale and nanoscale friction characteristics of micro-electro-mechanical-systems (MEMS) and nano-electro-mechanical-systems (NEMS) devices play an important role in determining performance, reliability, and lifetime of the overall system [14,15]. As such, controlling and minimizing friction and wear-related mechanical failures remains one of the greatest challenges for contemporary moving assembles. Nanoscale friction on graphene exhibits different behaviors from its 3D counterpart [16,17] because of its large surface-to-volume ratio, super-high mechanical stiffness, and strength [5]; all are highly desirable for wear protection. Most experimental studies have reported ultralow friction [18], or even superlubricity [19] sometimes accompanied by wear reduction [20]. The coefficient of friction for SiO_2 reduces from 0.68 to 0.12–0.22 after coating it with monolayer graphene [12]. Suspended graphene exhibits distinctive behavior in which nanoscale friction first increases and then decreases with increasing loads [21]. Deng et al. reported an adhesion-dependent negative friction coefficient on chemically modified graphite at the nanoscale [22]. Zhang et al. experimentally studied the nanoscale friction characteristics of graphene exfoliated onto weakly adherent silica substrates, and found that surface fluctuations are the main reason behind the suppression of thermal lubrication, which leads to an increase in friction force with temperature [23,24]. A scanning probe technique provides a clearer picture of friction at the nanometer scale, and an understanding of friction in layered materials [25]. The friction is attributable to the interaction between the incommensurate interface lattices [26], and is dominated by the so-called mechanism of "puckering" in front of the scanning tip, which increases the contact area and, therefore, the amount of friction [17]. Smolyanitsky et al. demonstrated that the experimentally observed reduction of friction with an increasing number of graphene layers in case of a narrow scanning tip can be a result of decreased sample deformation energy due to increased local contact stiffness under the scanning tip [27], and an increase of friction when the scanning tip is retracted away from the sample [28]. The JKR model [29] and DMT [30] model are generally successful in explaining friction behavior between smooth surfaces of various geometries, however, there is a lack of atomistic insight into the tip-sample interactions. So, detailed atomistic simulation has emerged as important

* Corresponding author.
E-mail addresses: wangweihuicool@163.com (W. Wang), victor_liu63@126.com (S. Liu).

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method to describe the nanoscale friction characteristics [16,27]. In brief, the factors including load [21], properties and roughness of substrate [31], scratch velocity [32,33], adhesion strength between contact surfaces [34], tip sizes [28] and temperature [35] significantly affect the friction properties of graphene or graphene-coated materials.

The number of graphene layers may be another important factor affecting the friction behaviors and characteristics of the overall system. Once more graphene layers are present, the interlayer van der Waals interaction can minimize the puckering effect and thereby reduce the friction. FFM (friction force microscopy) measurements found that monolayer epitaxial graphene on SiC exhibits higher friction than bilayer graphene [36]. Solution-processed graphene layers reduce friction and wear on steel surfaces in air [20]. However, there are few reports on the layer-dependent nanofriction of graphene coated systems [37]. The atomic characteristics are as of yet unexplored. Furthermore, it is especially interesting to investigate the friction properties of metal foils coated with graphene. Graphene grown on copper foils using the chemical vapor deposition method supplies the perfect test sample which shows great merit as a surface coating because of its excellent scalability and transferability [38]. In addition, there is no contamination and damage existing as there is no complex transfer process of graphene, such as PMMA (polymethyl methacrylate), which is typically used for the transfer process, that may have a significant effect on the results. The fundamental understanding of the atomistic process governing the friction behaviors and characteristics of multi-layer graphene/copper systems is still lacking.

In this study, we employ a faithful atomistic modeling to elucidate the friction behavior of graphene coated copper films, as well as the characteristics. The graphene/Cu system is selected because graphene on Cu (1 1 1) has a higher quality than that grown on other crystal orientations of copper [39]. We conduct atomic nano-indentation and scratching simulations of monolayer, bilayer, and trilayer graphene supported by a single crystal Cu (1 1 1) substrate. We have found that the friction reduces as the number of graphene layers increases. The corresponding behaviors and characteristics are explored in atomistic detail and excellent agreement with experiment is achieved.

2. Modeling and method

Our molecular dynamics (MD) calculations in this study are performed using the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [40]. We have used a rigid hemispherical diamond tip with a 2.5 nm radius to indent graphene layers grown on a Cu (1 1 1) substrate as shown in Fig. 1. Our model is smaller than that used in recent nano-indentation experiments, which has a size of 15.37 nm × 16.24 nm × 14.81 nm. However, our model dimension sizes are sufficiently large to simulate a semi-infinite boundary since the Cu (1 1 1) substrate block is cubic with a side length of around 5 times the tip radius [41] and the stress is localized and not impacted by the boundaries. In addition, the periodic boundaries are also employed in both lateral directions to avoid spurious edge effects. The choice above represents a compromise that minimizes finite-size effects while keeping the simulations affordable.

The quality of the MD simulation results significantly depends on the accuracy of the potential function used. The adaptive intermolecular reactive empirical bond order (AIREBO) potential [42] is used to describe the interatomic interaction for the tip and graphene, because it has been shown to accurately capture the bond-bond interaction between carbon and hydrogen atoms as well as bond breaking and bond reforming. Therefore, the AIREBO potential is also reliable for the studies of friction on graphene. The interaction cutoff distance in the switching function of the AIREBO potential must be selected carefully to avoid the known spurious post-hardening behavior arising from an improper cutoff distance. We set the cutoff distance to be 2.0 Å as suggested in [43]. Interatomic forces within the Cu (1 1 1) substrate are derived from an embedded atom method potential [44]. The interactions between tip and graphene, tip and copper, and graphene and copper are described by the van der Waals interaction as a 12–6 Lennard–Jones (LJ) potential. The parameters of the LJ potentials in our model are as follows: $\varepsilon_{C-C} = 2.86$ meV, $\sigma_{C-C} = 0.347$ nm, $\varepsilon_{C-Cu} = 11.7$ meV, $\sigma_{C-Cu} = 0.300$ nm [45], which actually indicates the adhesion strength between the tip, graphene, and substrate, a key factor in determining the frictional characteristics of the system.

The time increment of simulations is fixed at 1.0 fs, and a Langevin thermostat is applied to the whole system to maintain a stable temperature of 300 K. Firstly, the system is relaxed to minimum energy state for 15 ps. Then, the tip moves down and penetrates into the graphene/Cu (1 1 1) system at the speed of 0.05 nm/ps while the bottom copper atoms with thickness of 0.55 nm are fixed. Finally, the tip scratches laterally at a speed of 0.015 nm/ps while the normal (F_N) and lateral forces (F_L) acting on the rigid tip are recorded, which gives us much information of the scratch process of the tip on monolayer, bilayer, and trilayer graphene/Cu (1 1 1). Using the three procedures above, we can study the nanoscale friction behaviors and characteristics of different layers of graphene/Cu (1 1 1) systems by gradually changing the nano-indentation depth. In order to assess graphene's protective potential, analogous indentation and scratch simulations with a bare Cu (1 1 1) substrate are also performed.

3. Results and discussion

3.1. Scratching

A qualitatively distinct scratch behavior for a 2.06 nm nano-indentation depth on a monolayer graphene/Cu (1 1 1) system is presented in Fig. 2. After a nano-indentation depth of 2.06 nm,

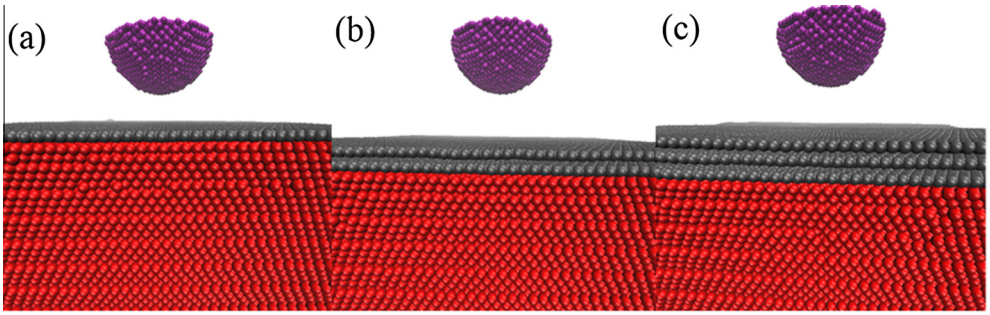

Fig. 1. Model of nano-indentation and scratch on (a) monolayer graphene/Cu (1 1 1), (b) bilayer graphene/Cu (1 1 1), and (c) trilayer graphene/Cu (1 1 1).