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# Frictional behavior of strained multilayer graphene: Tuning the atomic scale contact area

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ABSTRACT

Graphene is an ideal material to study nanoscale friction and modulating its frictional behavior is significant for the development of graphene-based micro- and nanoelectromechanical systems. Recently, strain engineering has been used to tune numerous properties of graphene. However few attentions have been paid to the strain effect on the frictional behavior of graphene. In this study, molecular dynamics simulations were carried out to study the friction coefficient of a strained multilayer graphene during scratching process under different scratch depths. It was found that the friction coefficient decreased under tensile strain while it increased under compressive strain. Further analysis of the atomic scale contact area was conducted to clarify the mechanism of such strain dependence. The results showed that the numbers of contacting atoms decreased as the strain increased from −8% to 8%, which was consistent with the friction coefficient variations under the applied strain. Therefore, the strain-dependent friction was attributed to the atomic scale contact area variations under the applied strain. This work shed light on the development of strain-based method to control nanoscale friction.



## 1. Introduction

As the development of micro- and nanoelectromechanical systems (MEMS/NEMS), tribological issues are demanding problems in such devices [1,2]. Therefore, controlling of friction is desirable at nanoscale. We have known for a long time that friction is proportional to the normal force and independent of the contact area [3]. Based on this understanding, a group of researchers developed methods to control nanoscale friction by tuning the normal force. Socoliuc et al. proposed an efficient method to control friction at the atomic scale by exciting the mechanical resonances of the sliding system perpendicular to the contact plane [4]. Pedraz et al. also found by atomic force microscopy (AFM) that the normal oscillations could be used to control the nanoscale friction and wear [5]. However, friction laws at nanoscale are found to be different from those at macroscale [6,7]. Mo et al.'s molecular dynamics (MD) simulations suggested that the nanoscale friction force depended linearly on the atomistic real contact area, which is determined by the interacting atoms [8]. Karuppiah et al. also found the dependence of nanoscale friction on the real contact area by AFM measurements [9]. Thus tuning the atomistic real contact area shed light on the development of new method to control friction at nanoscale. But how to effectively modulate the atomistic contact area is still a challenge.

Graphene, consisting of $sp^2$-bonded carbon atoms arranged in hexagon lattice, has received great attentions due to its remarkable properties [10–12]. Graphene is also an ideal material to study nanoscale friction owing to its two-dimensional structure [13–15]. Moreover, controlling its frictional behavior is crucial for graphene-based micro- and nanoelectromechanical devices. In the past few years, strain engineering of nano materials has opened new approach to modulate their properties [16,17]. The presence of mechanical strain was proved to have substantial effect on many properties of graphene. For instance, Moldovan et al. investigated the effect of mechanical strain on the electronic properties of graphene and found the applied strain could induce pseudo-magnetic field [18]. However, strain engineering of graphene has been studied extensively in terms of its electronic, magnetic and thermal properties [19,20]. Few attentions have been paid to the strain effect on the tribological properties of graphene. Recently, Kitt et al. [21] found the frictional behavior of few-layer graphene had strong dependence on the in-plane strain, which could smooth out the graphene and decrease the contact between graphene and substrate. Bai et al.'s MD simulations suggested that the frictional coefficient of a suspended graphene decreased with the increase of in-plane strain [22]. These studies provide new insights into ways of controlling nanoscale friction of graphene system.

In this study, for the purpose of exploring the effect of in-plane strain on the frictional behavior of multilayer graphene, molecular dynamics was adopted to simulate a diamond tip scratching on a strained four-

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layer graphene. The friction coefficients of the multilayer graphene were examined under different applied strains and scratch depths. Further analyses on the structure variations and atomistic real contact area during the scratching process were presented to derive the mechanism of the strain effect.

## 2. Methodology

MD simulations have been carried out by using LAMMPS software [23] to study the effect of external in-plane strain on the frictional behavior of multilayer graphene. The model system used in this study is illustrated in Fig. 1, where a hemispherical diamond tip with a radius of 25 Å was scratching on a pre-strained multilayer graphene with thickness of four layers. The interlayer spacing was 3.35 Å. The initial size of the graphene layer was 98 Å × 98 Å. In the simulation, the multilayer graphene was first strained along the *x*-axis by deforming the length of the graphene layer with a constant engineering strain rate of 0.01/ps. Then the system was relaxed for 50,000 steps to achieve minimal energy. After that, the scratching process was simulated. In the scratching process, the boundary atoms around the graphene layers were fixed to keep the position of the system. And the bottom graphene layer was also fixed, corresponding to a supported multilayer graphene. The rigid diamond tip was initially indented into the graphene with certain depth, and then scratched along the *x*-axis (in the same direction with the applied strain) with a constant speed of 5 m/s after another 50,000 steps of relaxation.

In the MD simulations, the intralayer atoms interactions of graphene and the interatomic interactions of diamond tip were described by the second generation of reactive empirical bond-order (REBO) potential [24]. The nonbonding interatomic interactions between the graphene atoms and diamond atoms were described by Lennard-Jones (LJ) potential with equilibrium distance of 3.4 Å [25,26]. The environmental temperature of the simulations was kept at 300 K controlled by the Berendsen thermostat method. All the simulations in the present study were performed in the constant volume constant internal energy (NVE) ensemble. The time steps for all the simulations were 1 fs, which is adequate for system relaxation by examining the stability through the root mean square deviations of the atoms. The duration for the scratching process was 600 ps with a distance of 30 Å.

## 3. Results and discussions

Based on the above described model, the scratching processes with different scratch depths were simulated under different applied strains. Due to our previous study [27,28], the scratch depth was defined as zero when the distance between the bottom of the diamond tip and the top layer of the graphene was 3.4 Å, since equilibrium distance of 3.4 Å was used in LJ potential. Fig. 2 shows the structures of the strained four-layer graphene under scratch depth of 5 Å at scratch distance of 15 Å. Here,

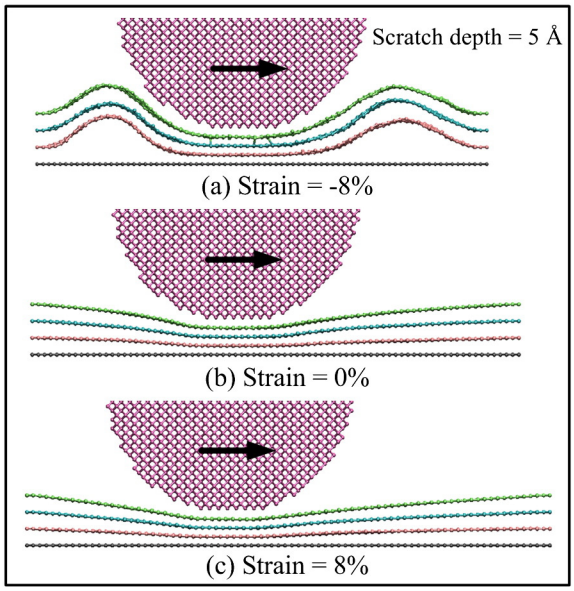


**Fig. 2.** Structures of the strained four-layer graphene under scratch depth of 5 Å at scratch distance of 15 Å: (a) −8%, (b) 0%, (c) 8%.

the negative sign of the applied strain refers to compressive strain and the positive sign refers to tensile strain. We can see that the diamond tip led to out-of-plane deformation of the graphene layers under all the applied strains. When the strain was −8%, large puckering deformation of the graphene layers took place around the diamond tip as shown in Fig. 2a. Moreover, there were a small amount of interlayer C—C bonds formed under strain of −8%, named cross-linking due to our previous study [28]. The formation of cross-linking resulted from the dangling bonds generated during the scratching process. In contrast, no cross-linking was observed when the applied strain was 0% and 8%, as shown in Fig. 2a and c.

Fig. 3 gives the friction coefficient variations of strained graphene during the scratching process under scratch depth of 5 Å. The friction force was calculated from the sum of the *x*-component forces acting upon all the atoms of the diamond tip. The friction coefficient was obtained by calculating the ratio of the friction force to the normal force acting on the diamond tip. As shown in Fig. 3, atomically resolved stick-slip behavior can be clearly observed under all the applied strains. The periodicity increased under tensile strain and decreased under compressive strain, which is in accordance with the variations of the lattice constant under the corresponding strain. The average friction coefficients were all nearly zero, indicating the graphene was in superlubricity state. However, it was obvious that the fluctuation amplitude of the friction coefficient changed under the applied strain.

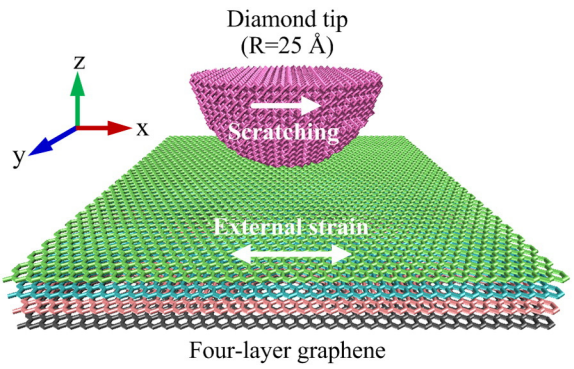


**Fig. 1.** Molecular dynamics simulation model of a diamond tip scratching on a strained four-layer graphene.

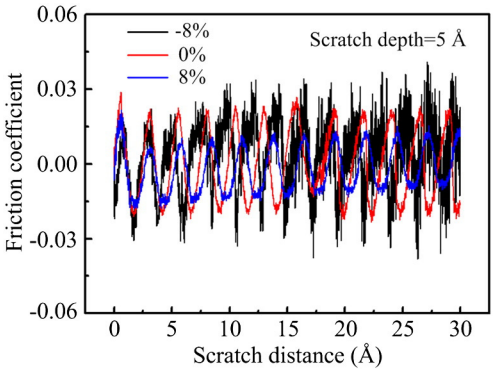


**Fig. 3.** Variations of the friction coefficient with the scratch distance under different strains (scratch depth = 5 Å).

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