

Origin of friction hysteresis on monolayer graphene

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Abstract: Load-dependent friction hysteresis is an intriguing phenomenon that occurs in many materials, where the friction measured during unloading is larger than that measured during loading for a given normal load. However, the mechanism underlying this behavior is still not well understood. In this work, temperature-controlled friction force microscopy was utilized to explore the origin of friction hysteresis on exfoliated monolayer graphene. The experimental observations show that environmental adsorbates from ambient air play an important role in the load dependence of friction. Specifically, the existence of environmental adsorbates between the tip and graphene surface gives rise to an enhanced tip–graphene adhesion force, which leads to a positive friction hysteresis where the friction force is larger during unloading than during loading. In contrast to positive friction hysteresis, a negative friction hysteresis where the friction force is smaller during unloading than during loading is observed through the removal of the environmental adsorbates upon *in situ* annealing. It is proposed that the measured friction hysteresis originates from the hysteresis in the contact area caused by environmental adsorbates between the tip and graphene. These findings provide a revised understanding of the friction hysteresis in monolayer graphene in terms of environmental adsorbates.

Keywords: friction hysteresis; friction force microscopy; graphene; environmental adsorbates; adhesion force

1 Introduction

A comprehensive and in-depth understanding of the frictional properties of two-dimensional (2D) materials, such as graphene, MoS₂, and h-BN, is of great significance for the development of practical 2D-material-based lubricants and coatings for friction reduction and wear protection applications [1–6]. In recent years, major progress has been achieved owing to the development of both friction force microscopy (FFM) [7] and atomistic simulation tools. It has been demonstrated that 2D materials possess numerous intriguing frictional properties [8–13]. For example, the friction on a 2D material surface was found to depend on the number of layers [8, 9]. In some cases, friction strengthening was also observed, where the

friction increased gradually during the initial motion and then leveled off [9, 12, 14]. It has also been reported that nanoscale friction on 2D materials can be strongly enhanced by chemical modification of the surface [15, 16]. Another interesting frictional property of 2D materials is the loading history dependence of friction [11, 17–19]. It was found that the friction force on graphene measured during unloading was significantly larger than that measured during loading for a given load [18, 19]. To explain this load-dependent friction hysteresis phenomenon, different mechanisms have been proposed. One suggestion is out-of-plane elastic deformation of the graphene surface [9, 11, 18]. The hysteresis was attributed to the relaxation of the local deformation of the graphene layer during unloading [11, 18]. Another hypothesis

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is the presence of a water meniscus at the tip–graphene contact area [19]. The hysteresis in the contact area caused by water leads to friction hysteresis [19].

The hysteresis in the contact area during a loading–unloading cycle was thought to originate from the adhesion force between the tip and surface of the 2D material, which suppressed the relaxation of the out-of-plane deformation of the sample. In other words, the friction hysteresis of 2D materials may be affected by the properties of the material surface. It was shown that exposing the graphite surface to ambient air could enhance the adhesion between the tip and graphite [11]. When the tip slides against the graphite surface, the topmost graphene layer delaminates from the graphite, which results in a significant increase in friction as the applied normal load is decreased (negative friction coefficient). Additionally, airborne hydrocarbons and other small molecules inevitably adsorb on the surfaces of 2D materials when exposed to ambient air, which could affect their surface properties [20–25]. It has been demonstrated that airborne contaminants can cause the hydrophilic surface of 2D materials to become hydrophobic [21, 24]. Similarly, the frictional behaviors of 2D materials can be affected by hydrocarbon contamination [26–29]. For instance, environmental adsorbates can self-assemble into highly oriented nanoscale stripes on the graphene surface, leading to friction anisotropy on monolayer graphene [27, 28].

Previous studies have demonstrated that the presence of water molecules at the sliding interface can influence the load-dependent friction hysteresis [19, 30]. Nevertheless, the effect of adsorbed hydrocarbon molecules from ambient air on friction hysteresis remains unclear. In this study, the role of environmental adsorbates on the load-dependent friction hysteresis of exfoliated monolayer graphene deposited on a SiO₂/Si substrate was explored by utilizing temperature-controlled FFM. In contrast to the friction hysteresis that has been reported where the friction force measured during unloading was larger than that measured during loading, an inverted friction hysteresis was investigated where the friction measured during unloading was smaller than that measured during loading upon *in situ* annealing. The experimental results measured at a relative humidity (RH) of 40%

indicated that the water molecules were not the reason for the inverted friction hysteresis. In combination with the friction hysteresis on bulk graphite, it is proposed that the observed friction hysteresis on monolayer graphene originated from the high adhesion force as a result of the adsorption of environmental adsorbates between the tip and graphene.

2 Materials and methods

Monolayer graphene was prepared by mechanical cleavage of natural graphite flakes (Alfa Aesar, USA) under ambient conditions (25 °C; RH ~35%). The graphene flakes deposited on the SiO₂/Si substrate were observed using an optical microscope (Olympus BX53, Japan) before the friction measurement. The friction and topography images of graphene were measured using a Cypher ES atomic force microscope (Asylum Research, USA). For the friction measurement, a commercial atomic force microscope cantilever (AC240TS-R3, Olympus) with a resonance frequency of ~70 kHz and a spring constant of ~2.0 N·m⁻¹ was employed. The lateral sensitivity of the cantilever was calculated using a noncontact method [31, 32]. For the annealing treatment of graphene, a cooler/heater was used to control the temperature, and a humidity detector was employed to obtain the RH of the test environment.

3 Results and discussion

FFM friction measurements conducted on exfoliated monolayer graphene supported on a SiO₂/Si substrate are summarized in Fig. 1. Although the topography image shows a uniform monolayer of graphene (Fig. S1 in the Electronic Supplementary Material (ESM)), the simultaneously recorded friction force image clearly reveals the existence of distinct frictional domains on the graphene layer (Fig. 1(a)). It has been reported that ripple distortions in monolayer graphene induced by residue stress result in anisotropic friction, and the friction anisotropy-driven domains can be removed through thermal annealing treatment as a result of the disappearance of residual stress [10]. In another report, it was proposed that the frictional domains originate from environmental adsorbates

that can form highly oriented nanostripes on the surface of monolayer graphene [27]. To determine the reason for the observed frictional domains in the experiment in the present study, the monolayer graphene was first annealed *in situ* at 100 °C for 20 min under argon protection. The friction contrast in the domains on graphene disappeared after the annealing treatment (Fig. 1(b)). Then, the monolayer graphene was exposed to ambient air for 10 min, and the frictional domains appeared again (Fig. 1(c)). The nanostripes on the monolayer graphene were also observed through high-resolution imaging (Fig. 1(d)), and the width of each stripe was approximately 4 ± 0.2 nm (Fig. 1(f), black curve), which is consistent with previous reports [27, 28]. No detectable features were observed for the annealed graphene, as shown in Figs. 1(e) and 1(f). It has been reported that environmental adsorbates are mainly small hydrocarbon molecules (e.g., alkanes, alkenes, and alcohols) [24, 28]. In addition, because the adsorption of hydrocarbon molecules on the surface of graphene mainly occurs through physical adsorption, they can desorb upon

annealing treatment [21]. Therefore, the repeatable and reversible phenomenon of frictional domains, combined with the observed nanostripes on the graphene, confirmed the existence of environmental adsorbates on the surface of the exfoliated monolayer graphene in the experiment.

To further explore the influence of environmental adsorbates on the frictional behavior of monolayer graphene, the friction of monolayer graphene was measured under different normal loads. As shown in Fig. 2(a), for a given normal load, the frictional force on the as-prepared graphene measured during unloading was larger than that measured during loading (positive friction hysteresis), which is consistent with previous reports [18, 19]. Then, the graphene sample was annealed *in situ* at 100 °C for 20 min in an atomic force microscopy (AFM) environmental chamber under the protection of argon and then cooled to room temperature for the friction measurement. It was observed that the friction hysteresis on graphene was inverted, as shown in Fig. 2(b), where the friction force measured during the loading process was larger than that

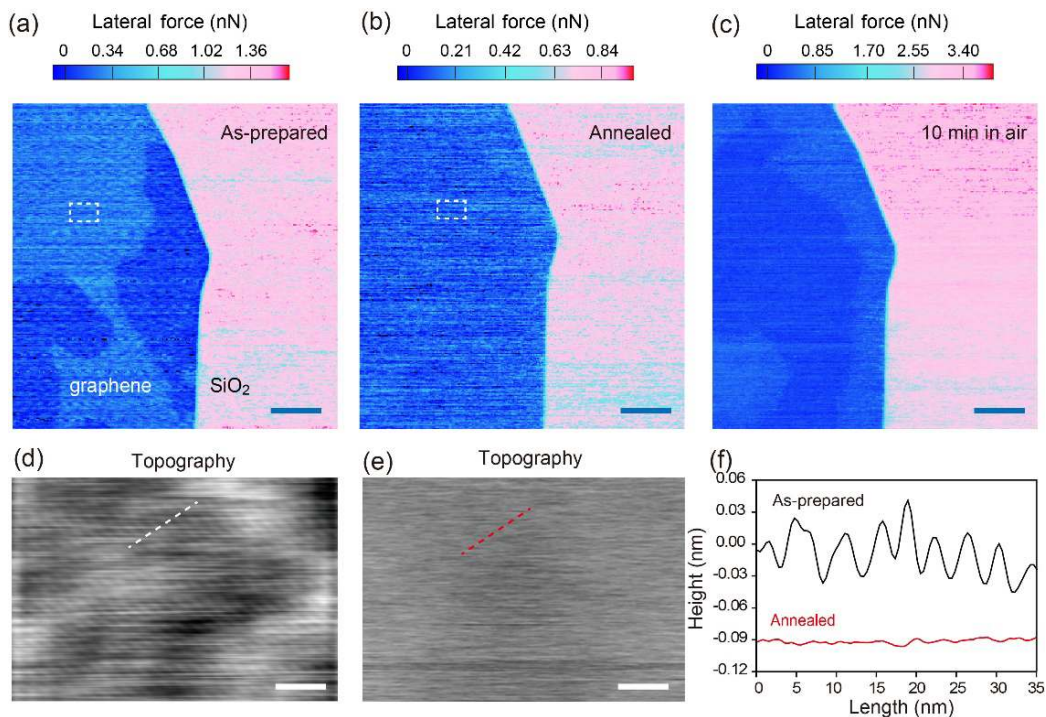


Fig. 1 Friction force map and high-resolution topography image of graphene. (a) Friction force map of the as-prepared graphene, (b) the graphene after *in situ* annealing at 100 °C for 20 min, and (c) the annealed graphene after exposure to ambient air for 10 min. Scale bars are 1 μ m. (d) and (e) High-resolution topography image of the as-prepared graphene surface and annealed graphene surface taken within the dashed white rectangle marked in (a) and (b), respectively. Scale bars are 20 nm. (f) The height line profiles along the dashed lines marked on (d) and (e), respectively.

measured during the unloading process (negative friction hysteresis). To the best of our knowledge, negative friction hysteresis in monolayer graphene has not been reported previously. When friction measurements were performed at high relative humidity (RH 40%) after the annealing treatment, inverted friction hysteresis was still observed (Fig. 2(c)). However, the friction hysteresis returned to the positive state after the graphene was exposed to ambient air for 10 min (Fig. 2(d)). In addition, the loads corresponding to the tip detached from the graphene surface during the process of unloading, defined herein as the slide-off force, were approximately -15 , 10 , 12 , and -40 nN for as-prepared, annealed, RH 40%, and re-exposed graphene, respectively. The slide-off force is regarded as a type of adhesion between the tip and graphene, which is related to the sliding history [33]. The variation in the slide-off force could indicate the influence of adsorbates on the interaction between the tip and graphene after a loading–unloading cycle. A smaller sliding-off force means that the tip is easier to detach from the surface of graphene after the annealing process has been conducted. In addition, the interactions between the tip and graphene are composed of two forces: the adhesion force and normal

load (compressive force). The normal loads alone are not sufficient to maintain contact between the tip and graphene when the adhesion force becomes smaller, which could cause the tip to detach from the graphene surface, even at a normal load force (e.g., 10 nN for annealed graphene and 12 nN for RH 40% graphene). Furthermore, the frictional force measured during the loading or unloading process for a given normal load was found to decrease from the as-prepared graphene to annealed graphene.

To identify whether the negative friction hysteresis was caused by the drifting of the normal load signal, the friction hysteresis experiment was repeated. As shown in Fig. S2 in the ESM, the friction hysteresis phenomenon was successfully replicated, indicating that this phenomenon was not an accidental event. It is easy to understand that the friction changes if the normal load signal drifts.

To exclude the influence of normal load signal drifting on the experimental results of friction, the following experiment was conducted. For the annealed monolayer graphene, the friction was measured under three different normal loads. Under each normal load, the tip continued to slide against the surface of the graphene for 15 min, which was equivalent to the

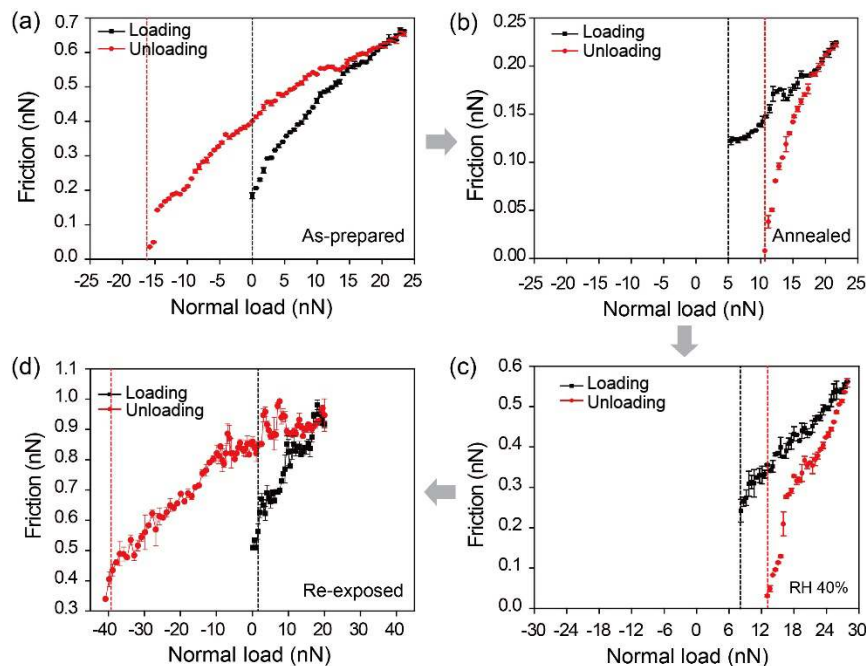


Fig. 2 Evolution process of friction hysteresis. (a) Normal load dependence of friction measured on the as-prepared graphene, (b) the graphene after *in situ* annealing at 100 °C for 20 min, (c) the annealed graphene exposing to high relative humidity (RH 40%), and (d) the annealed graphene re-exposed to ambient air for 10 min.

time used in a loading–unloading cycle. As shown in Fig. S3(a) in the ESM, there is no significant change in friction under each normal load, which means that the positive pressure applied to the sample surface by the probe is stable. The corresponding normal load versus time data also clarified that no obvious normal load signal drifting occurred (Fig. S3(b) in the ESM).

The adhesion force between the atomic force microscope tip and graphene is an important factor that can influence the frictional behaviors of graphene [11, 34]. Therefore, the adhesion force between the tip and graphene, specifically referring to the pull-off force, was quantified by measuring the conventional force–separation curves [35]. The pull-off force could reveal the initial interaction between the tip and the graphene without sliding. For each type of graphene, 400 force curves were recorded to obtain the statistical distribution values of the adhesion forces. The experimental results are shown in Figs. 3(a)–3(d). The adhesion forces between the tip and as-prepared, annealed, RH 40%, and re-exposed graphene were approximately 5.32 ± 0.12 , 4.58 ± 0.25 , 5.25 ± 0.31 , 5.76 ± 0.30 nN, respectively. It is clear that the adhesion force decreased after the annealing treatment (Fig. 3(e)), which was a tendency similar to the trend of friction,

indicating that the decrease in adhesion force is one of the reasons for the decrease in friction.

Fundamentally, the adhesion force (F_{ad}) between the tip and graphene consists of four parts, which can be expressed as

$$F_{ad} = F_{vdw} + F_{me} + F_{el} + F_{ch} \quad (1)$$

where F_{vdw} is the van der Waals force, F_{me} is the meniscus force, F_{el} is the electrostatic force, and F_{ch} is the chemical bonding force. Considering that it is difficult to form chemical bonds between the tip and graphene, F_{ch} can be neglected. In addition, F_{el} can be inconsiderable because there are no voltage-induced charges on the tip and the graphene. Here, F_{vdw} is a distance-dependent force between atoms (molecules), which can also be affected by the number and type of atoms (molecules) [36]. The annealing process removed the environmental adsorbates on the surface of the graphene (Figs. 1(b) and 1(f)) and decreased the RH of the AFM sample chamber (Fig. S4 in the ESM), which resulted in decreased F_{vdw} and F_{me} values. In short, the annealing treatment decreased the adhesion force between the tip and graphene, which originated from the removal of environmental adsorbates and the reduction of water molecules. Next, whether the

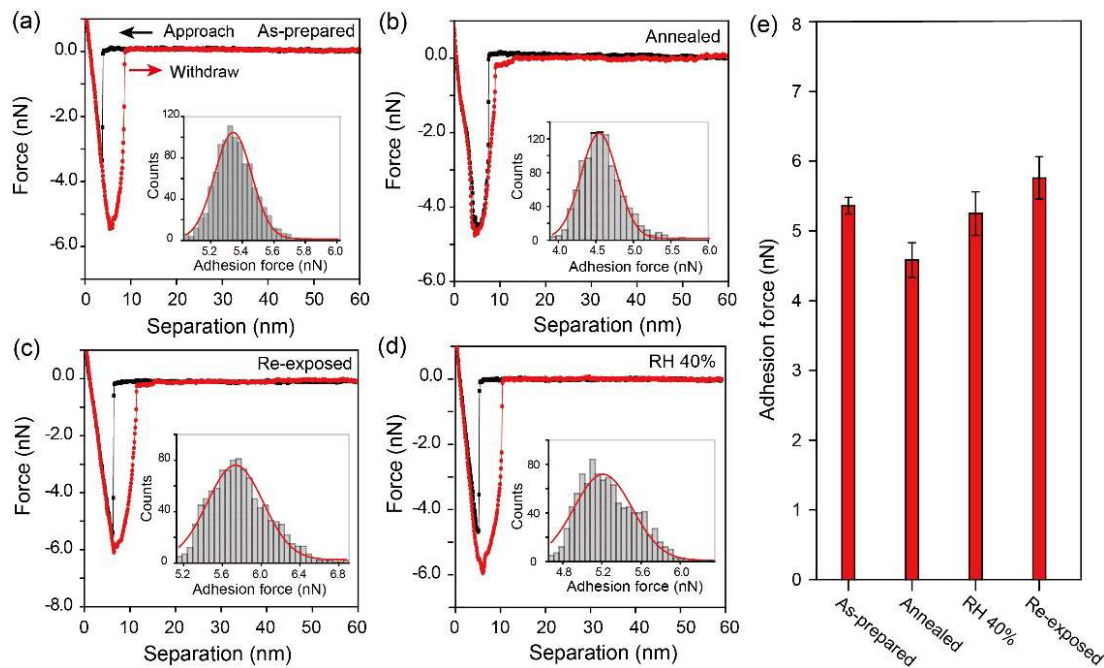


Fig. 3 Adhesion force between the tip and graphene. Utilizing the force curves to measure the adhesion force between the tip and (a) as-prepared, (b) annealed, (c) RH 40%, and (d) re-exposed graphene. Four hundred force curves were recorded under each condition to obtain the statistical value of adhesion force. (e) Adhesion forces between the tip and graphene obtained from (a–d).

variation of water molecules leads to negative friction hysteresis was investigated. The friction of annealed graphene at an RH of 40%, which is equivalent to recovering only the water molecules between the tip and graphene, was measured. As shown in Fig. 2(c), the friction force was larger than that of annealed graphene at a given normal load, and the phenomenon of inverted friction hysteresis still existed, showing that the changes of water molecules are one reason for the variation in friction and not the reason for the negative friction hysteresis.

To explore further the contribution of the environmental adsorbates to friction hysteresis, load-dependent friction measurements on bulk graphite were conducted. As shown in Figs. 4(a) and S5, nanostructured structures were also observed on the surface of the bulk graphite, with a typical width of approximately 4 nm (Fig. 4(b)). Then, the friction on bulk graphite was measured without any treatment. The friction hysteresis was not investigated, as shown in Fig. 4(c). The friction hysteresis of the annealed bulk graphite remained the same (Fig. 4(d)). Although no evolution process of friction hysteresis was observed on the bulk graphite, the variation of friction force on bulk graphite and the tip-sample sliding-off force after the annealing

process is consistent with the phenomenon observed on monolayer graphene. The same experiment was also performed on the annealed bulk graphite. The experimental results are presented in Fig. S6 in the ESM, which also indicates that the normal load signal is stable during the friction measurement process.

It has been demonstrated that monolayer graphene adhering to the sliding tip can generate out-of-plane deformation owing to the small bending stiffness of the graphene sheet, which results in increased contact area and friction between the tip and graphene [9]. Based on the evolution of friction hysteresis experiments conducted on SiO₂/Si supported monolayer graphene, it is concluded that the friction hysteresis originated from the assembly of environmental adsorbates, which gave rise to a high tip-graphene adhesion force. During the loading process, the contact area between the tip and graphene increases with increasing load, whereas, during the unloading process, the structure of graphene tends to return to its initial state, leading to a decreased contact area. However, the high adhesion force caused by the environmental adsorbates leads to the hysteresis of the contact area between the tip and graphene, which results in friction hysteresis. As shown in Figs. 5(a)–5(c), as the friction measurement

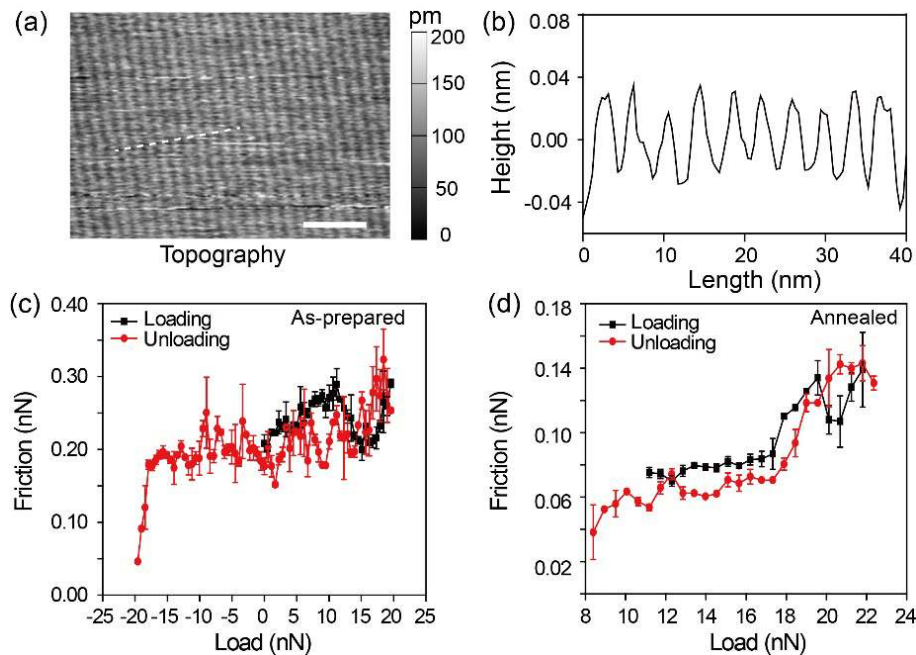


Fig. 4 Friction hysteresis of bulk graphite. (a) High-resolution topography image of bulk graphite surface, observing the oriented stripes. The scale bar is 20 nm. (b) The height line profile along the white dashed line marked on (a). (c) Friction hysteresis of as-prepared bulk graphite and (d) the same sample after annealed at 100 °C for 20 min.

was conducted, the environmental adsorbates on the as-prepared graphene surface accumulated between the tip and graphene, leading to an enhanced adhesion force. The enhanced adhesion force was reflected in the slide-off force between the tip and graphene during unloading, as shown in Fig. 2. Through the annealing treatment, the environmental adsorbates on the surface of the graphene could be removed (Figs. 5(g)–5(j)). In comparison with the as-prepared graphene, the contact area between the tip and annealed graphene decreased for a given normal load as the adhesion force decreased because of the removal of the environmental adsorbates. During the unloading process, for the as-prepared graphene, the high adhesion force owing to the environmental adsorbates suppressed the graphene returning to its original state. The high adhesion force during unloading

caused a larger contact area than that during loading for a given normal load, which leads to a positive friction hysteresis (Figs. 5(d)–5(f)). However, for the annealed graphene, the adhesion force between the tip and graphene was not high enough to suppress the trend of graphene deformation recovery, which resulted in a smaller contact area during unloading than that during loading for a given normal load, leading to negative friction hysteresis (Figs. 5(j)–5(l)).

For bulk graphite, because the interlayer interaction between graphene layers is much greater than the adhesion force between the tip and graphene, it is very difficult to create out-of-plane deformation, regardless of the presence of environmental adsorbates. Therefore, there was no evolution of friction hysteresis on the bulk graphite as the annealing process was conducted.

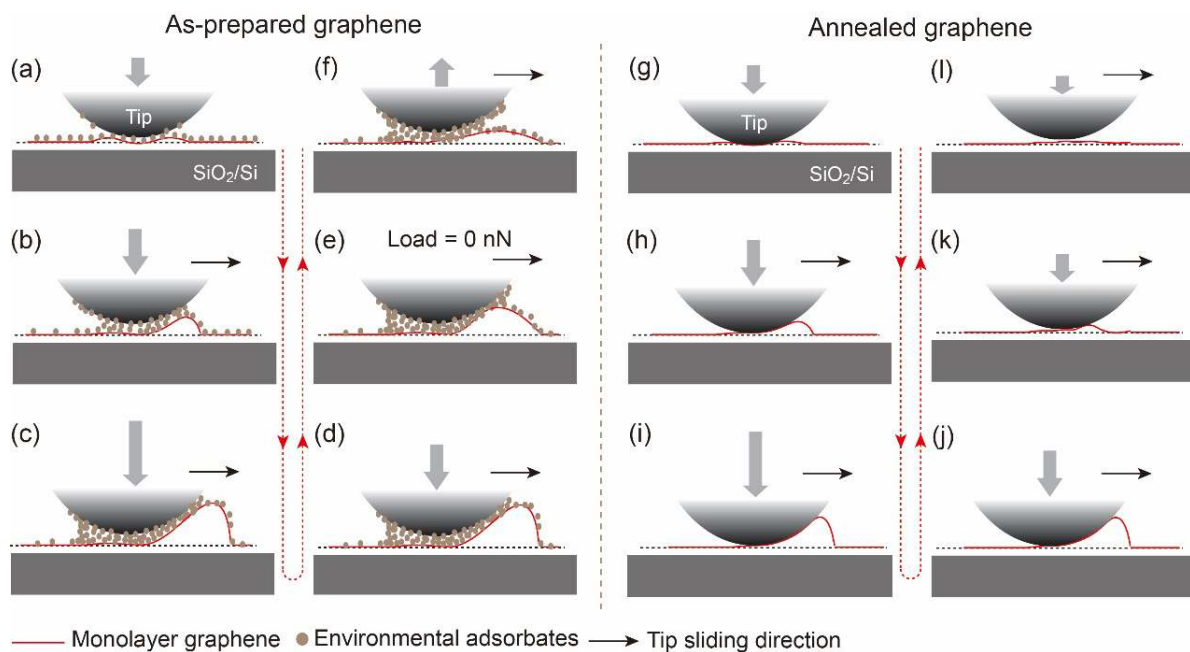


Fig. 5 Schematic illustration of the load-dependent friction hysteresis on SiO₂/Si supported graphene. The red solid line depicts the structure of monolayer graphene during the load-dependent friction characterization. The black dashed line depicts the initial state of graphene, and the brown solid circle depicts the environmental adsorbates. For the as-prepared graphene, (a) the structure of graphene begins to deform as the tip contacts the graphene, and the environmental adsorbates at this stage are distributed on the graphene surface. (b) As the tip starts to slip, an out-of-plane deformation is created and the environmental adsorbates begin to accumulate between the tip and graphene. (c) As the load increases and environmental adsorbates accumulate, the height of the out-of-plane deformation is enhanced. (d) When the load begins to decrease after reaching a maximum load, the high tip–graphene adhesion force suppresses the recovery of the out-of-plane deformation. At zero load (e) and even at adhesion force (f), the out-of-plane deformation is still visible. For the annealed graphene, the environmental adsorbates were removed. Out-of-plane deformation is also observed, and the height increases as the normal load increases (h, i). As the load decreases, the adhesion force between the tip and graphene is not high enough to inhibit the recovery of the out-of-plane deformation (j, k). The structure of graphene almost returned to its initial state and separated from the tip (l), even when the load still existed. The length of the vertical gray arrow indicates the amount of load applied on the graphene. The upward arrow illustrated in (f) indicates that only adhesion force exists.

4 Conclusions

In conclusion, the influence of environmental adsorbates on the friction hysteresis of exfoliated monolayer graphene supported on a SiO₂/Si substrate was explored through temperature-controlled FFM. For the first time, the transition from positive friction hysteresis (friction being larger during unloading than during loading) to negative friction hysteresis (friction being smaller during unloading than during loading) through the removal of the environmental adsorbates was observed. According to the experimental results, it was proposed that the existence of environmental adsorbates between the tip and graphene during the loading process can enhance the tip–graphene adhesion force. The high adhesion force suppressed the recovery of the out-of-plane deformation to its original state, which resulted in a larger contact area during unloading than during the loading process for a given normal load. However, for the annealed graphene, there were no environmental adsorbates on the graphene surface. Thus, the adhesion force between the tip and graphene was not sufficiently high to suppress the recovery of the graphene deformation. In other words, the contact area between the tip and annealed graphene during unloading was smaller than that during loading, which caused negative friction hysteresis. No friction hysteresis was observed on the bulk graphite owing to the strong interlayer interaction of graphite, which suppressed the out-of-plane deformation of the topmost layer of graphite. The findings reveal that environmental adsorbates play a significant role in the friction properties of 2D materials. The results also suggest that it is necessary to ensure surface cleanliness when 2D materials are used as solid lubricants in nanoscale applications.

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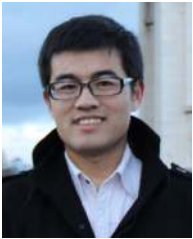
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