Friction **8**(2): 462–470 (2020)
https://doi.org/10.1007/s40544-019-0288-0
ISSN 2223-7690
CN 10-1237/TH
RESEARCH ARTICLE

Temperature and velocity dependent friction of a microscale graphite–DLC heterostructure

Yujie GONGYANG[1,2], Wengen OUYANG[3,*], Cangyu QU[1,2], Michael URBAKH[3,4], Baogang QUAN[5,6], Ming MA[7,8,*], Quanshui ZHENG[1,2,8]

[1] *Department of Engineering Mechanics, Tsinghua University, Beijing 100084, China*

[2] *Center for Nano and Micro Mechanics, Tsinghua University, Beijing 100084, China*

[3] *School of Chemistry and The Sackler Center for Computational Molecular and Materials Science, Tel Aviv University, Tel Aviv 69978, Israel*

[4] *The Raymond and Beverly Sackler Faculty of Exact Sciences, Tel Aviv University, Tel Aviv 69978, Israel*

[5] *Beijing National Laboratory for Condensed Matter Physics and Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*

[6] *School of Physical Sciences, University of Chinese Academy of Sciences, Beijing 100190, China*

[7] *State Key Laboratory of Tribology, Tsinghua University, Beijing 100084, China*

[8] *Department of Mechanical Engineering, Tsinghua University, Beijing 100084, China*

Received: 11 January 2019 / Revised: 28 February 2019 / Accepted: 11 March 2019

Abstract: One of the promising approaches to achieving large scale superlubricity is the use of junctions between existing ultra-flat surface together with superlubric graphite mesas. Here we studied the frictional properties of microscale graphite mesa sliding on the diamond-like carbon, a commercially available material with a ultra-flat surface. The interface is composed of a single crystalline graphene and a diamond-like carbon surface with roughness less than 1 nm. Using an integrated approach, which includes Argon plasma irradiation of diamond-like carbon surfaces, X-ray photoelectron spectroscopy analysis and Langmuir adsorption modeling, we found that while the velocity dependence of friction follows a thermally activated sliding mechanism, its temperature dependence is due to the desorption of chemical groups upon heating. These observations indicate that the edges have a significant contribution to the friction. Our results highlight potential factors affecting this type of emerging friction junctions and provide a novel approach for tuning their friction properties through ion irradiation.

Keywords: friction; graphite; diamond like carbon; irradiation; desorption

1 Introduction

Graphite has been used as a solid lubricant and known to show ultralow friction since ancient times [1]. With the development of nanotribology over the past 20 years, a new intriguing frictional property of this old material was discovered, termed as superlubricity. The relatively strong intralayer bonding and weak interlayer interaction lead to almost frictionless sliding at the incommensurate contact between single crystalline graphite surfaces, i.e., to the state called superlubricity [2]. This idea was firstly predicted theoretically [3, 4] and later confirmed by experiments with nanoscale graphitic contacts under restricted environments such as dry nitrogen [5] and ultrahigh vacuum [6]. Considering the practical importance of having a frictionless interface, it is easy to understand the great efforts which have been made during the last decades to achieve robust superlubricity on larger scale and under ambient conditions [7–17]. An important step in this direction has been made recently, when so-called self-retraction experiments with graphite

* Corresponding authors: Wengen OUYANG, E-mail: wengenouyang@mail.tau.ac.il; Ming MA, E-mail: maming16@tsinghua.edu.cn

mesas provided a direct evidence of superlubricity in micrometer graphitic contacts [7]. It was shown that the ultralow friction observed in these experiments relies on using atomically smooth, single crystalline structure of the mesa surfaces placed in an incommensurate contact.

To widen the scope of possible applications of the microscale superlubricity, the frictional properties of graphite mesa in contact with other materials should be investigated. In fact, microscale heterojunctions of graphite and other materials including *h*-BN [18], MoS_2 [19–21], mica [22], diamond-like carbon [23, 24] have been fabricated by transferring graphite mesas onto the target surface. These heterostructures open wide opportunities for tuning interfacial frictional properties. Diamond-like carbon (DLC), which is widely used as coating material due to its unique mechanical, optical, electrical, chemical, and tribological properties [25–28] is a promising candidate for transfer target. The sizes of DLC films can easily reach macroscopic scale while remaining very smooth [29]. Thus, studies of the tribological property of the graphite-DLC heterojunctions are of particular interest. Investigations of the temperature and velocity dependence of friction may provide significant insights into the mechanisms of sliding friction [18, 30–33], however such measurements have not yet been carried out for graphite/DLC heterojunctions.

In addition to studies of velocity and temperature dependence of friction, investigations of the effect of irradiation on friction may provide an essential information on mechanisms of sliding friction, since this method allows to tune the chemistry and structure of surfaces at atomic scale in a controllable way. It is usually considered that irradiation causes a disorder on surfaces, however recent experiments on electron or ion irradiation demonstrated that graphitic systems exhibit a unique self-healing phenomenon, where a fast diffusion of carbon atoms in the layered structure leads to healing the defects introduced by irradiation [34, 35]. Thus it would be interesting to investigate the effects of irradiation on the sliding friction in the graphite/DLC heterojunction.

In this paper, we investigated the temperature and velocity dependence of friction for microscale graphite/DLC heterojunction. The interface is composed of a single crystalline graphene and DLC surfaces with roughness less than 1 nm. The logarithmic dependence of friction on velocity observed in our experiments is consistent with the mechanism of thermally activated friction, and the temperature dependence of friction is explained using a Langmuir adsorption model, which describes adsorption and desorption of gas molecules on the edges of graphite mesa. It is found that Argon plasma irradiation can effectively tune the frictional properties of the heterojunctions. X-ray photoelectron spectroscopy (XPS) characterizations revealed that Argon plasma irradiation removes the oxygen from the graphite edges, and converts part of sp^2 carbon states into the sp^3 states.

2 Experimental procedures

Graphite/DLC heterojunctions were fabricated using the technique developed in a previous publication [23]. Briefly, square shaped graphite mesas (typical height of 1 μm and side length of 4 μm) were fabricated through electron beam lithography and reactive ion etching (RIE). The so-called self-retraction phenomenon reported in former publications [8] allowed us to select self-retractable graphite mesas with atomically smooth and single crystalline contact surfaces. The self-retractable graphite mesas were then transferred onto a DLC surface using a tungsten tip attached to a nano-manipulator (Kleindiek MM-3A) to form the heterojunctions under ambient conditions. The DLC surface was prepared in the same way as reported in a previous publication [23], i.e. by cutting a DLC slice without lubricant from a hard disk drive (manufactured by the West Digital Corporation) into smaller pieces with sizes of 1 cm × 1 cm. Here we used the *a*-C:N type DLC, for which the sp^3/sp^2 ratio is 56% and I_D/I_G of Raman spectrum is 0.64.

After transferring the graphite mesa on DLC, the temperature and velocity dependencies of friction in the heterojunctions were investigated using an atomic force microscope (AFM) (NT-MDT) with a heating stage and an environmental chamber, as shown in Fig. 1(a). The sample was first annealed at 150 ℃ and then cooled down naturally to room temperature. To keep the relative humidity at low level (<5%), dry N_2 was pumped into the chamber throughout the annealing and the measurement processes. To measure the friction of heterojunctions at different temperatures,

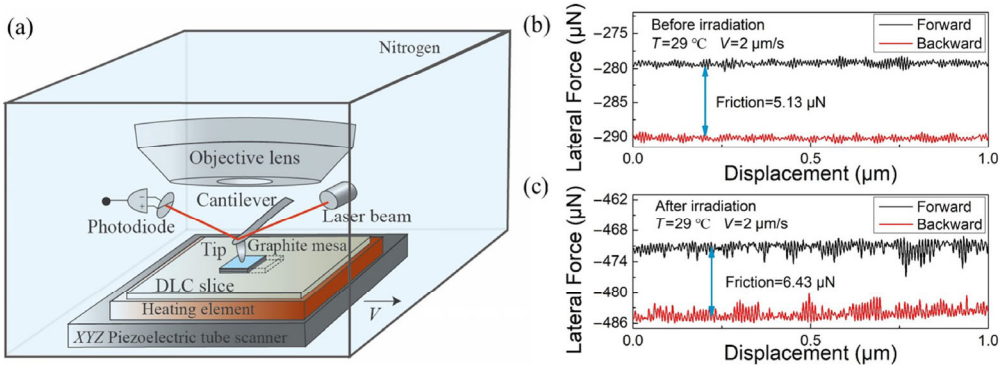


Fig. 1 (a) Schematic experimental setup. The graphite/DLC heterojunction was mounted on the AFM with a heating stage and an environment chamber filled with N_2. The friction was measured by sliding the graphite mesa against the DLC surface using the AFM tip; typical lateral force versus displacement curves for mesas before (b) and after (c) Argon plasma irradiation. The average friction force is calculated as $F = (f_{\text{Forward}} - f_{\text{Backward}})/2$.

the samples were heated to the target temperature T (ranging from room temperature to 150 ℃, in a random order) and equilibrated for 30 minutes before measuring friction. Then the graphite mesa was pressed by the AFM tip from the top, with a typical normal load of 56.5 μN. Under such a normal load, the static friction at the interface between the AFM tip and the mesa top surface was usually larger than the friction at the mesa-DLC interface. Thus, when the AFM tip was scanned laterally with a speed V, the mesa slide against the DLC surface, and the corresponding lateral force was measured. The average friction force is calculated as $F = (f_{\text{Forward}} - f_{\text{Backward}})/2$, as shown in Fig. 1(b).

Similar friction measurements were also conducted with graphite mesas exposed to Argon plasma irradiation. For these mesas, the experimental process was the same except that Argon plasma irradiation with energy of 300 eV and beam density of 0.5 mA/cm^2 was used to irradiate the edges of graphite mesa for a duration of 30 seconds before transferring the mesa onto the DLC surface. The typical lateral force versus displacement curves for mesas obtained after Argon plasma irradiation of DLC are similar to those for mesas without Argon plasma treatment, as shown in Fig. 1(c). The measurements corresponding to Figs. 1(b) and 1(c) were both performed at temperature of T = 29 ℃ with sliding velocity of V = 2 μm/s.

3 Experimental results

For all systems studied at T = 150 ℃ before irradiation we found a friction coefficient of about 0.008, indicating a superlubric state of graphite/DLC heterojunctions. The velocity dependencies of friction force measured for temperatures ranging from room temperature to 150 ℃ are plotted in Fig. 2. For all investigated temperatures the presented results show a logarithmic increase of friction with velocity v. Such observations are consistent with the oretic predictions of Prandtl-Tomlinson (PT) model of thermally activated friction [36, 37]. Similar velocity dependencies of friction have been previously found in scanning probe microscope

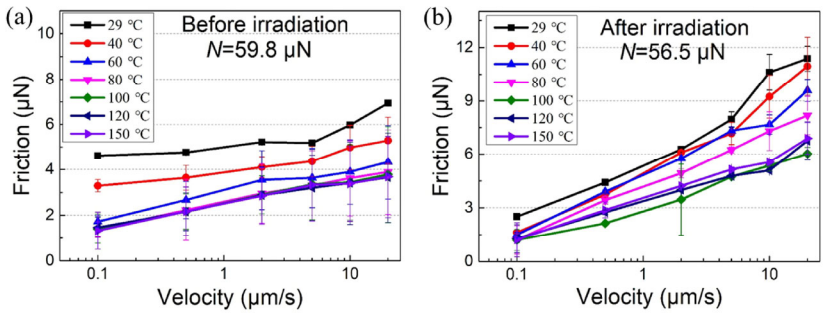


Fig. 2 Velocity dependence of friction at different temperature for mesas not exposed (a) and exposed (b) to Argon plasma irradiation. The slope of the logarithmic dependence of friction on velocity increases after irradiation.

experiments, where the frictional contacts are usually in the nanoscale range [38, 39]. Here we report that the friction of microscale graphite/DLC heterojunctions show logarithmic velocity dependence, suggesting thermally activated sliding mechanism. Moreover, our experimental results show that the slope of the logarithmic dependence increases by about 120% after the Argon plasma irradiation, indicating an increase of sliding energy barriers. The reason will be explained below.

While the velocity dependence of friction of graphite/DLC heterojunctions follows predictions of conventional thermally activated model of friction, its temperature dependence cannot be described by this model. Figure 3(a) shows that for mesas not exposed to irradiation the friction force decreases with temperature for $T < 100$ °C, and then saturates as temperature increases ($T > 100$ °C). The frictional behavior of mesas after Argon plasma irradiation is similar, but the data are more scattered (see Fig. 3(b)). The effect of water evaporation on friction can be excluded since N_2 atmosphere was kept throughout the experiments, and annealing at 150 °C was performed before all experiments.

To understand the effect of temperature on friction, XPS measurements were conducted, using the same experimental procedures as described in Ref. [23]. Briefly, since the size of the XPS spot is typically much larger than the size of graphite mesas, a direct characterization of graphite mesa edges is difficult. To solve this problem, we fabricated a larger graphite sample (with size of 1 cm) following exactly the same procedures for the graphite mesa fabrication. The side surface of this graphite sample was characterized by XPS both before and after the Argon plasma irradiation treatment. Thus, we believe that the XPS characterization of these graphite samples reasonably reflects properties of graphite mesa edges before and after the Argon plasma treatment.

The XPS spectra of the graphite edges before and after irradiation are shown in Fig. 4(a), peaks corresponding to O1s (532.7 eV), N1s (398.5 eV), and C1s (284.8 eV) can be identified in the spectrum before the irradiation, while after the irradiation, the N1s peak disappears and the height of O1s peak decreases. More specifically, it is shown that the fraction of oxygen drops from 18.9% to 0.53% after irradiation. Interestingly, the previous publication [23] has shown that the reactive ion etching process used for the fabrication of graphite mesa produces a considerable amount of oxygen at graphite edges. Here we found that the oxygen can be efficiently removed by the Argon plasma irradiation. Analyzing the components of the C1s peak, we estimated the relative fraction of carbon atoms in sp^3 and sp^2 states, as shown in Figs. 4(b) and 4(c). The one with a binding energy of 284.5 eV corresponds to the C–C sp^2 bonds in the graphitic network, and the peak at 285.1 eV is attributed to the carbon bonds with sp^3 hybridization. The two peaks with binding energy equal to 286.5 eV and 288.3 eV correspond to C–O and O–C=O bonds, respectively [40–42]. It should be noted that the ratio of sp^3 carbon to sp^2 carbon increases from 1.41 to 2.73 after irradiation. Before the irradiation, a considerable amount of C–O and C=O bonds were found, whereas after the irradiation the amount of C–O bonds reduces significantly and no evidence of presence of C=O bonds are found. Overall, our results suggest that Argon plasma irradiation removes most of the oxygen from the graphite edge and converts part of the sp^2

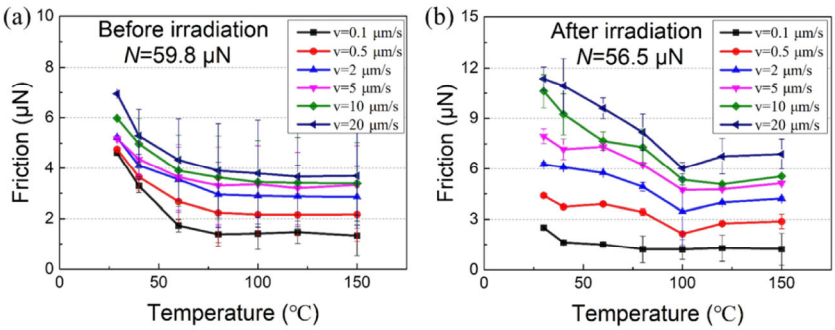
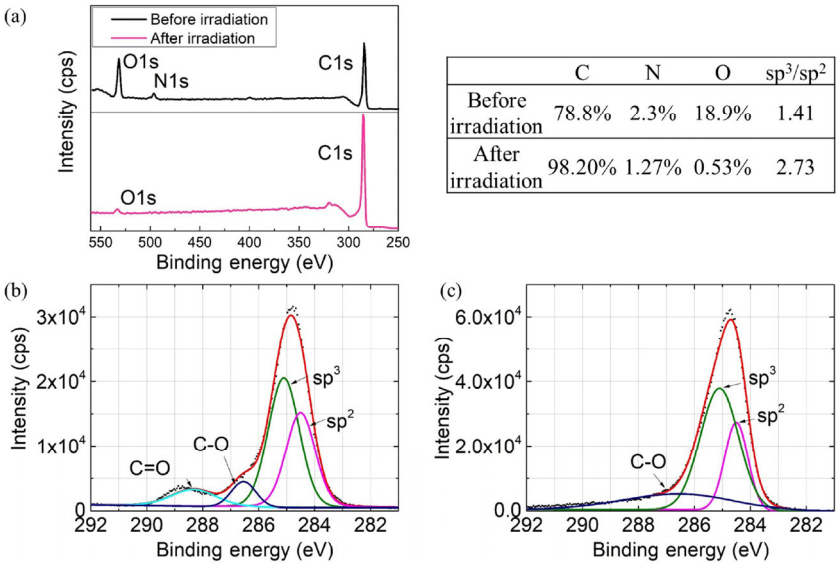


Fig. 3 Temperature dependence of friction at different velocity for mesas not exposed (a) and exposed (b) to Argon plasma irradiation. Friction decreases at low temperatures, and then levels with increasing temperature.



	C	N	O	sp^3/sp^2
Before irradiation	78.8%	2.3%	18.9%	1.41
After irradiation	98.20%	1.27%	0.53%	2.73

Fig. 4 XPS data for the graphite mesa edges. (a) XPS spectra of graphite edge before and after Argon plasma irradiation, and table showing a percentage of elements present at the edges. C1s spectra of the graphite edge before (b) and after (c) irradiation.

carbon into sp^3 carbon. The former effect leads to the reduction of the friction while the latter one enhances it due to the higher fraction of hydrocarbons. The competing effects of the two different chemical changes on friction lead to larger scattering in friction after irradiation as shown in Fig. 3.

4 Model simulations

Based on the XPS analysis shown above, we may conclude that significant changes in friction of graphite/DLC heterojunctions, which are induced by irradiation, may result from the effect of irradiation on the chemical structure of the graphite edges. Thus, the temperature dependence of friction may be caused by the adsorption and desorption of the chemical groups at the graphite mesa edges. Since the experiments were conducted under N_2 atmosphere, the amount of adsorbed molecules is very low. Under these conditions, it is reasonable to assume that the friction force is proportional to the amount of molecules adsorbed at the edges [43, 44]. To estimate the effect of temperature on the amount of adsorbed molecules, we used a classical Langmuir adsorption model for two types of molecules [24, 45–47]. The adsorbates which could be present at the mesa edges include H_2 released from DLC under heating, hydroxyl groups exhibited by the XPS spectra and hydrocarbons which are inevitably present.

For the i^{th} (i = 1, 2) type of adsorbate, its adsorption and desorption rates can be expressed as follows [47–49]:

$$k^i_{\text{ads}} = p_i \cdot k^0_{\text{a}} \exp(-E^i_{\text{ads}} / k_{\text{B}}T), \quad (1)$$

$$k^i_{\text{des}} = k^0_{\text{d}} \exp(-E^i_{\text{des}} / k_{\text{B}}T), \quad (2)$$

where E^i_{ads}, E^i_{des}, and p_i are the adsorption energy barrier, desorption energy barrier, and the partial pressure of the i^{th} type of adsorbate. k^0_a and k^0_d are prefactors, k_{B} is the Boltzman constant, and T is the absolute temperature.

Based on the absolute rate theory (ART), the kinetics of adsorption/desorption processes can be captured by the Wigner-Polanyi expression for the rate of adsorption [49]:

$$\frac{\mathrm{d}\theta_i}{\mathrm{d}t} = k^i_{\text{ads}}\left(1 - \sum_{i=1}^{2}\theta_i\right) - k^i_{\text{des}}\theta_i \ (i = 1, 2), \quad (3)$$

here θ_i is the fractional surface coverage of the i^{th} type of adsorbate, t is time. It should be noted that in Eq. (3) we extended the original form to be suitable for multi-type of adsorbates. At equilibrium, $\frac{\mathrm{d}\theta_i}{\mathrm{d}t} = 0$ (i = 1, 2), we can easily get the Langmuir isotherm from Eq. (3) as,

$$\theta_{\text{tot}} = \frac{\sum_{i=1}^{2} p_i r_{\text{ad}}^i}{1+\sum_{i=1}^{2} p_i r_{\text{ad}}^i}, \quad (4)$$

here, $\theta_{\text{tot}} = \theta_1 + \theta_2$ is the total coverage. $r_{\text{ad}}^i = \text{e}^{\frac{Q_i}{k_B T}} (i = 1,2)$ is the ratio between the rate of adsorption and desorption and $Q_i = E_{\text{des}}^i - E_{\text{ads}}^i (i = 1,2)$ is the adsorption heat for the i^{th} type of adsorbate.

As discussed above, the friction is proportional to the number of adsorbates at the edges of graphite mesa, thus to reveal its intrinsic dependence on temperature, the friction is normalized to its value at T = 302 K as $f^* = f/f\,(302\text{ K})$ for different sliding velocities. We use Eq. (4) to fit the normalized friction force.

To reveal the effect of temperature on friction, we show in Fig. 5 the temperature dependence of the normalized friction forces (symbols), where the forces measured for different velocities were normalized to their values found for T = 302 K. Then, the upper and lower bounds of normalized experimental data were fitted to the total adsorbate coverage calculated according to Eq. (4). The fitting of experimental data obtained before the Argon plasma irradiation yields $p_1 = 4.17\times10^{-4} - 1\times10^{-3}$, $p_2 = 1\times10^{-9} - 2.99\times10^{-8}$, and $Q_1 = 0.01 - 0.0334$ eV, $Q_2 = 0.309 - 0.353$ eV. We note that for both upper and bottom bounds the estimated adsorption heats Q_1 and Q_2 match the literature values for adsorption heat of H_2 on graphite [50] and hydroxyl groups at the edges of graphite mesa [51]. After the Argon plasma irradiation, the fitting yields $p_1 = 1.04\times10^{-4} - 4.77\times10^{-3}$, $p_2 = 2.99\times10^{-8} - 1.18\times10^{-7}$, and $Q_1 = 0.0154 - 0.0336$ eV, $Q_2 = 0.22 - 0.231$ eV, where Q_1 and Q_2 match the literature values for adsorption heats of H_2 and hydrocarbon on graphite [50]. The absence of hydroxyl groups after irradiation is consistent with our XPS characterization, which shows that Argon plasma irradiation reduces the amount of oxygen. This analysis, shows that the levelling off of the friction force observed with increasing temperature occurs as a result of desorption of hydroxyl groups (before irradiation) and hydrocarbon molecules (after irradiation), while the H_2 molecules remain adsorbed by the edges.

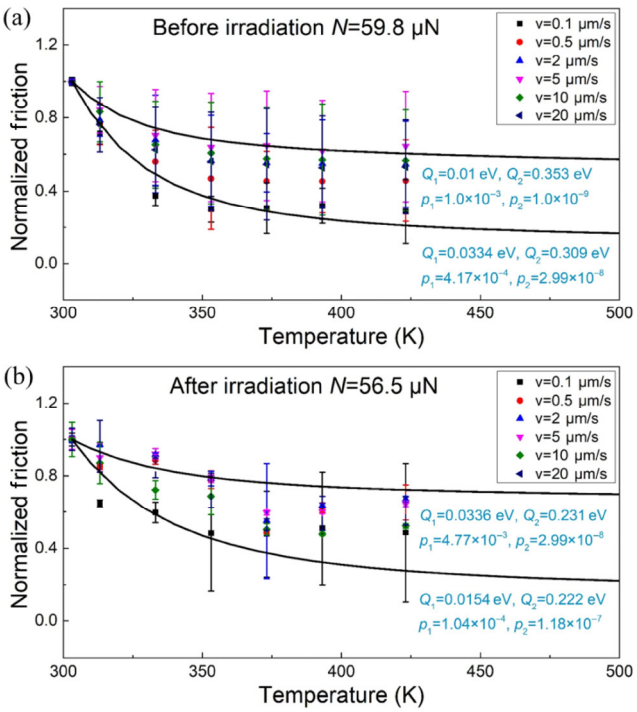


Fig. 5 Temperature dependence of the normalized friction forces (symbols) measured for different velocity for mesas before (a) and after (b) Argon plasma irradiation. The solid curves are fitting results according to Eq. (4).

5 Conclusions

In conclusion, by investigating the temperature and velocity dependence of friction for microscale graphite/DLC heterojunctions, we found that the velocity dependence follows the thermally activated sliding mechanism, whereas the observed temperature dependence cannot be explained based on existing theories. To reveal the mechanism of friction in these heterojunctions, we further studied the effects of Argon plasma irradiation on friction. Using XPS analysis of graphite mesas and Langmuir adsorption model, we conclude that the observed temperature dependence of friction is determined by the desorption of chemical groups from the edges of graphite mesa that occurs with increasing temperature, which largely affect the frictional properties. Our results provide insights into the origin of frictions in the graphite/DLC heterojunctions, which have the potential for practical applications, and offer a viable approach for tuning the frictional properties via Argon plasma irradiation.

Acknowledgements

Wengen Ouyang acknowledges the financial support from a fellowship program for outstanding postdoctoral

researchers from China and India in Israeli Universities. Ming Ma wishes to acknowledge the financial support by Thousand Young Talents Program and the NSFC grant Nos. 11632009, 11772168, and 11890673. Quanshui Zheng wishes to acknowledge the financial support by the NSFC grant No. 11890671.

References

[1] Spreadborough J. The frictional behaviour of graphite. *Wear* **5**(1): 18–30 (1962)

[2] Hod O, Meyer E, Zheng Q S, Urbakh M. Structural superlubricity and ultralow friction across the length scales. *Nature* **563**(7732): 485–492 (2018)

[3] Aubry S. Exact models with a complete devil's staircase. *J Phys C-Solid State Phys* **16**(13): 2497–2508 (1983)

[4] Hirano M, Shinjo K. Atomistic locking and friction. *Phys Rev B* **41**(17): 11837–11851 (1990)

[5] Dienwiebel M, Verhoeven G S, Pradeep N, Frenken J W M, Heimberg J A, Zandbergen H W. Superlubricity of graphite. *Phys Rev Lett* **92**(12): 126101 (2004)

[6] Dietzel D, Ritter C, Mönninghoff T, Fuchs H, Schirmeisen A, Schwarz U D. Frictional duality observed during nanoparticle sliding. *Phys Rev Lett* **101**(12): 125505 (2008)

[7] Zheng Q S, Jiang B, Liu S P, Weng Y X, Lu L, Xue Q K, Zhu J, Jiang Q, Wang S, Peng L M. Self-retracting motion of graphite microflakes. *Phys Rev Lett* **100**(6): 067205 (2008)

[8] Liu Z, Yang J R, Grey F, Liu J Z, Liu Y L, Wang Y B, Yang Y L, Cheng Y, Zheng Q S. Observation of microscale superlubricity in graphite. *Phys Rev Lett* **108**(20): 205503 (2012)

[9] Yang J R, Liu Z, Grey F, Xu Z P, Li X D, Liu Y L, Urbakh M, Cheng Y, Zheng Q S. Observation of high-speed microscale superlubricity in graphite. *Phys Rev Lett* **110**(25): 255504 (2013)

[10] Wang W, Dai S Y, Li X D, Yang J R, Srolovitz D J, Zheng Q S. Measurement of the cleavage energy of graphite. *Nat Commun* **6**: 7853 (2015)

[11] Zhang R F, Ning Z Y, Zhang Y Y, Zheng Q S, Chen Q, Xie H H, Zhang Q, Qian W Z, Wei F. Superlubricity in centimetres-long double-walled carbon nanotubes under ambient conditions. *Nat Nanotechnol* **8**(12): 912–916 (2013)

[12] Liu S W, Wang H P, Xu Q, Ma T B, Yu G, Zhang C H, Geng D C, Yu Z W, Zhang S G, Wang W Z, et al. Robust microscale superlubricity under high contact pressure enabled by graphene-coated microsphere. *Nat Commun* **8**: 14029 (2017)

[13] Zheng X H, Gao L, Yao Q Z, Li Q Y, Zhang M, Xie X M, Qiao S, Wang G, Ma T B, Di Z F, et al. Robust ultra-low-friction state of graphene via moire superlattice confinement. *Nat Commun* **7**: 13204 (2016)

[14] Kawai S, Benassi A, Gnecco E, Söde H, Pawlak R, Feng X L, Müllen K, Passerone D, Pignedoli C A, Ruffieux P, et al. Superlubricity of graphene nanoribbons on gold surfaces. *Science* **351**(6276): 957–961 (2016)

[15] Cihan E, Ipek S, Durgun E, Baykara M Z. Structural lubricity under ambient conditions. *Nat Commun* **7**: 12055 (2016)

[16] Berman D, Deshmukh S A, Sankaranarayanan S K R S, Erdemir A, Sumant A V. Macroscale superlubricity enabled by graphene nanoscroll formation. *Science* **348**(6239): 1118–1122 (2015)

[17] Li J J, Li J F, Luo J B. Superlubricity of graphite sliding against graphene nanoflake under ultrahigh contact pressure. *Adv Sci* **5**(11): 1800810 (2018)

[18] Song Y M, Mandelli D, Hod O, Urbakh M, Ma M, Zheng Q S. Robust microscale superlubricity in graphite/hexagonal boron nitride layered heterojunctions. *Nat Mater* **17**(10): 894–899 (2018)

[19] Wang L F, Zhou X, Ma T B, Liu D M, Gao L, Li X, Zhang J, Hu Y Z, Wang H, Dai Y D, et al. Superlubricity of a graphene/MoS_2 heterostructure: A combined experimental and dft study. *Nanoscale* **9**(30): 10846–10853 (2017)

[20] Liu Y M, Song A S, Xu Z, Zong R L, Zhang J, Yang W Y, Wang R, Hu Y Z, Luo J B, Ma T B. Interlayer friction and superlubricity in single-crystalline contact enabled by two-dimensional flake-wrapped atomic force microscope tips. *Acs Nano* **12**(8): 7638–7646 (2018)

[21] Zhou X, Jin K, Cong X, Tan Q H, Li J Y, Liu D M, Luo J B. Interlayer interaction on twisted interface in incommensurate stacking MoS_2: A raman spectroscopy study. *J Colloid Interface Sci* **538**: 159–164 (2019)

[22] Liu B T, Wang J, Peng X N, Qu C Y, Ma M, Zheng Q S. Direct fabrication of graphite-mica heterojunction and *in situ* control of their relative orientation. *Mater Des* **160**: 371–376 (2018)

[23] Gongyang Y J, Qu C Y, Zhang S M, Ma M, Zheng Q S. Eliminating delamination of graphite sliding on diamond-like carbon. *Carbon* **132**: 444–450 (2018)

[24] Bhowmick S, Banerji A, Alpas A T. Friction reduction mechanisms in multilayer graphene sliding against hydrogenated diamond-like carbon. *Carbon* **109**: 795–804 (2016)

[25] Grill A. Diamond-like carbon: State of the art. *Diam Relat Mater* **8**(2–5): 428–434 (1999)

[26] Erdemir A, Donnet C. Tribology of diamond-like carbon films: Recent progress and future prospects. *J Phys D*: *Appl Phys* **39**(18): R311 (2006)

[27] Erdemir A, Eryilmaz O L, Fenske G. Synthesis of diamondlike carbon films with superlow friction and wear properties. *J Vac Sci Technol A* **18**(4): 1987–1992 (2000)

[28] Erdemir A, Eryilmaz O. Achieving superlubricity in DLC films by controlling bulk, surface, and tribochemistry. *Friction* **2**(2): 140–155 (2014)

[29] Peng X L, Barber Z H, Clyne T W. Surface roughness of diamond-like carbon films prepared using various techniques. *Surf Coat Technol* **138**(1): 23–32 (2001)

[30] Vu C C, Zhang S M, Urbakh M, Li Q Y, He Q C, Zheng Q S. Observation of normal-force-independent superlubricity in mesoscopic graphite contacts. *Phys Rev B* **94**(8): 081405 (2016)

[31] Wang Z J, Ma T B, Hu Y Z, Xu L, Wang H. Energy dissipation of atomic-scale friction based on one-dimensional prandtl-tomlinson model. *Friction* **3**(2): 170–182 (2015)

[32] Barel I, Urbakh M, Jansen L, Schirmeisen A. Multibond dynamics of nanoscale friction: The role of temperature. *Phys Rev Lett* **104**(6): 066104 (2010)

[33] Barel I, Urbakh M, Jansen L, Schirmeisen A. Temperature dependence of friction at the nanoscale: When the unexpected turns normal. *Tribol Lett* **39**(3): 311–319 (2010)

[34] Krasheninnikov A V, Banhart F. Engineering of nanostructured carbon materials with electron or ion beams. *Nat Mater* **6**(10): 723–733 (2007)

[35] Ma M D, Liu J Z, Wang L F, Shen L M, Xie L, Wei F, Zhu J, Gong Q M, Liang J, Zheng Q S. Reversible high-pressure carbon nanotube vessel. *Phys Rev B* **81**(23): 235420 (2010)

[36] Sang Y, Dubé M, Grant M. Thermal effects on atomic friction. *Phys Rev Lett* **87**(17): 174301 (2001)

[37] Dudko O K, Filippov A E, Klafter J, Urbakh M. Dynamic force spectroscopy: A fokker-planck approach. *Chem Phys Lett* **352**(5–6): 499–504 (2002)

[38] Jansen L, Höelscher H, Fuchs H, Schirmeisen A. Temperature dependence of atomic-scale stick-slip friction. *Phys Rev Lett* **104**(25): 256101 (2010)

[39] Gnecco E, Bennewitz R, Gyalog T, Loppacher C, Bammerlin M, Meyer E, Güntherodt H J. Velocity dependence of atomic friction. *Phys Rev Lett* **84**(6): 1172–1175 (2000)

[40] Siokou A, Ravani F, Karakalos S, Frank O, Kalbac M, Galiotis C. Surface refinement and electronic properties of graphene layers grown on copper substrate: An XPS, UPS and eels study. *Appl Surf Sci* **257**(23): 9785–9790 (2011)

[41] Paik N. Raman and XPS studies of DLC films prepared by a magnetron sputter-type negative ion source. *Surf Coat Technol* **200**(7): 2170–2174 (2005)

[42] Mérel P, Tabbal M, Chaker M, Moisa S, Margot J. Direct evaluation of the sp^3 content in diamond-like-carbon films by XPS. *Appl Surf Sci* **136**(1–2): 105–110 (1998)

[43] Ma M, Sokolov I M, Wang W, Filippov A E, Zheng Q S, Urbakh M. Diffusion through bifurcations in oscillating nano- and microscale contacts: Fundamentals and applications. *Phys Rev X* **5**(3): 031020 (2015)

[44] Ouyang W G, de Wijn A S, Urbakh M. Atomic-scale sliding friction on a contaminated surface. *Nanoscale* **10**(14): 6375–6381 (2018)

[45] Hanaor D A H, Ghadiri M, Chrzanowski W, Gan Y X. Scalable surface area characterization by electrokinetic analysis of complex anion adsorption. *Langmuir* **30**(50): 15143–15152 (2014)

[46] Connor P A, McQuillan A J. Phosphate adsorption onto TiO_2 from aqueous solutions: An in situ internal reflection infrared spectroscopic study. *Langmuir* **15**(8): 2916–2921 (1999)

[47] House J E. *Principles of Chemical Kinetics*. Boston (USA): Academic Press, 2007.

[48] Rudzinski W, Panczyk T. Kinetics of isothermal adsorption on energetically heterogeneous solid surfaces: A new theoretical description based on the statistical rate theory of interfacial transport. *J Phys Chem B* **104**(39): 9149–9162 (2000)

[49] Clark A. *The Theory of Adsorption and Catalysis*. New York (USA): Academic Press, 1970.

[50] Grinev I V, Zubkov V V, Samsonov V M. Calculation of isosteric heats of molecular gas and vapor adsorption on graphite using density functional theory. *Colloid J* **78**(1): 37–46 (2016)

[51] Klomkliang N, Nantiphar O, Thakhat S, Horikawa T, Nakashima K, Do D D, Nicholson D. Adsorption of methanol on highly graphitized thermal carbon black effects of the configuration of functional groups and their interspacing. *Carbon* **118**: 709–722 (2017)

Yujie GONGYANG. She received her bachelor degree in engineering mechanics in 2014 from Beijing Institute of Technology, Beijing, China. After then, she studied for her doctorate in the Department of Engineering Mechanics at Tsinghua University. Her research interests include nano/micro mechanics and superlubricity.

Wengen OUYANG. He received his Ph.D. degree in engineering mechanics from Tsinghua University, China, in 2016. Currently, he is a postdoc in School of Chemistry of Tel Aviv University, Israel. His research interests include nanotribology, superlubricity, and developing realistic interlayer force fields for 2D materials.

Ming MA. He received his Ph.D. degree in engineering mechanics from Tsinghua University, China, in 2011. He joined the State Key Laboratory of Tribology at Tsinghua University from 2016. His current position is an associate professor. His research areas cover nanotribology, nanofluidics, superlubricity, and diffusion on surfaces or under confinement.