Controlling, and in many cases minimizing, friction is a goal that has long been pursued in history. From the classic Amontons–Coulomb law to the recent nanoscale experiments, the steady-state friction is found to be an inherent property of a sliding interface, which typically cannot be altered on demand. In this work, we show that the friction on a graphene sheet can be tuned reversibly by simple mechanical straining. In particular, by applying a tensile strain (up to 0.60%), we are able to achieve a superlubric state (coefficient of friction nearly 0.001) on a suspended graphene. Our atomistic simulations together with atomically resolved friction images reveal that the in-plane strain effectively modulates the flexibility of graphene. Consequently, the local pinning capability of the contact interface is changed, resulting in the unusual strain-dependent frictional behavior. This work demonstrates that the deformability of atomic-scale structures can provide an additional channel of regulating the friction of contact interfaces involving configurationally flexible materials.

Results

To characterize the frictional behavior of graphene with different in-plane strains, monolayer graphene samples were prepared by mechanical exfoliation and transferred to SiO2/Si substrates with prefabricated circular holes (SI Appendix, Fig. S1). The graphene samples were first placed in an autoclave for 2 to 7 d to be equilibrated with a preset high pressure. Then the graphene samples were taken out in ambient conditions and they would bulge due to the pressure difference across the graphene layer (21–24). By adjusting the pressure difference, various graphene samples with different tensile strains could be obtained and their surface topography and friction were measured via atomic force microscopy (AFM) as schematically shown in Fig. 1A.

Fig. 1B shows 4 typical 3D topographic images of monolayer graphene with different strains (the corresponding line profiles can be found in SI Appendix, Fig. S1). The relative strain of each graphene sample was characterized by Raman spectroscopy as shown in Fig. 1C. When no external pressure difference was imposed, the graphene appeared to be flat with a slight sink-in due to the sidewall adhesion. The Raman G band and 2D band of the sample are located around 1581 and 2674 cm−1, respectively. In contrast, the graphene samples with an external pressure difference would bulge out, appearing like a bubble showing an obvious shift in the peak position of the Raman G band and

Significance

It has long been conjectured that the steady-state friction of an elastic contact is an inherent property of the sliding interface, which depends on the contacting materials and typically cannot be altered on demand. In this work, we demonstrate that the friction on a graphene sheet can be actively modulated by in-plane straining. In particular, by applying a tensile strain (up to 0.60%), we are able to further reduce the surface friction of monolayer graphene to a superlubricating state. This unusual effect is attributed to the changes in the atomic contact quality of the sliding interface. Our work demonstrates the very example, where the atomic-scale interfacial interactions can be directly regulated via macroscopic operations.
where $\Delta \varepsilon$ is the relative strain, $\gamma = 1.8$ is the Grüneisen parameter, $\omega$ and $\omega_0$ are the Raman frequencies of the G band at the strained state and the reference state, respectively. In the experiments, we chose the flat graphene sample without an external pressure difference as the reference state. It should be noted that, although the graphene appears to be flat in topography, it might still have a finite tensile strain due to the sidewall adhesion (26, 27). Based on the reference state, the relative strains at the center of the graphene samples shown in Fig. 1C are 0, 0.05, 0.14, and 0.37%, respectively.

Frick measurements were carried out in the central area of each graphene sample to minimize the potential impact of the curvature effect (more discussions can be found in SI Appendix, Fig. S2). In addition to the suspended samples, we also measured the friction of the SiO$_2$ supported graphene, which was previously found to be loosely bonded with intrinsic ripples (17, 28, 29). Fig. 1D shows mean friction versus normal load curves for both the supported graphene and the suspended graphene samples. As indicated in Fig. 1D, friction of the suspended graphene exhibits a substantial reduction compared to the supported case. Moreover, the friction of the suspended graphene decreases monotonically with increasing tensile strain and it gradually levels off at high strains. By fitting the friction force as a linear function of the normal load, we could obtain the effective coefficients of friction (COFs) for graphene with different strains. As shown in Fig. 1E, the effective COF of graphene with 0.37% relative tensile strain is only 0.0013, which is ~6 times lower than the supported graphene and is in the regime of superlubricity based on the level of COF (30). It should be noted that in order to exclude the potential influence of the systematic change in the tip shapes, the friction tests were conducted in a random sequence (in the order of 0.14%, 0.37%, flat, supported, and 0.05%). We also performed repetitive friction measurements and confirmed that the above trend was qualitatively reproducible (details can be found in SI Appendix, Fig. S3) regardless of choice of specific samples or probes.

To further investigate the friction modulation effect by strain, we carried out atomic-scale friction measurements on supported graphene and suspended graphene with relative strains up to 0.60%. As shown by the representative friction loops in Fig. 2A, all friction signals show the clear atomic-scale stick–slip. For the supported graphene, the apparent friction strengthening can be observed, i.e., the lateral force increases initially with sliding and remains at a steady state after a few atomic periods. This evolution effect is consistent with the previous reports and is attributed to the transient behavior of the confined graphene sheet (16, 19). In contrast, for the suspended graphene, the evolution behavior is absent and only the regular atomic-scale stick–slip is observed. In the meantime, when the tensile strain increases, the difference between the trace and the retrace force signals decrease, indicating less energy dissipation. The reduction in energy dissipation is accompanied by a reduction in the local peak lateral force as shown in Fig. 2A. To quantify the differences, we calculated the local peak force and the energy dissipation per unit period during the steady state, and plotted them in Fig. 2B. One can clearly see that as the strain increases, both the energy dissipation and the local peak lateral force decrease significantly, consistent with the larger-scale results shown in Fig. 1. In addition to the variation in energy dissipation, the atomically resolved friction data in Fig. 2A also suggest that the asymmetry between the stick and the slip phases gradually becomes weaker when the tensile strain increases (see more details in SI Appendix, Fig. S4). According to Socoliuc et al. (31), the reduction in stick-and-slip asymmetry indicates a diminishing energy corrugation.
and a transition from the stick–slip to continuous sliding state. To quantify the transition behavior, we estimated the parameter, \( \eta \), based on the experimental data in Fig. 2A. The values of \( \eta \) were estimated to be 4.5 \pm 0.4, 2.8 \pm 0.6, 2.1 \pm 0.6, 1.8 \pm 0.6, and 1.4 \pm 0.4 for the supported, flat, 0.14\%, 0.37\%, and 0.6\% cases, respectively. Therefore, the slip is a nearly continuous sliding state when the graphene strain reaches 0.6\%. Using the peak lateral force of the stick–slip curves shown in Fig. 2A, we also estimated the friction shear strength for graphene samples with different strains. As shown in SI Appendix, Fig. S5, the friction shear strength decreases from 0.1 to 0.06 MPa as the strain increases from 0 to 0.6\%. Because the frictional behavior of graphene can be affected by surface adhesion (32, 33), we conducted adhesion force microscopy using AFM. The 2D adhesion maps as well as the normalize adhesion forces are shown in Fig. 2C. Similar to previous findings (33), the overall adhesion on the suspended graphene is generally smaller (up to \( \sim 10\% \)) than on the supported case; however, there is no significant difference among the suspended graphene with different strains. Therefore, the significant friction modulation on graphene via straining is unlikely to originate from the difference in surface adhesion.

Discussion

To explore the mechanism of friction tuning via straining, molecular dynamics (MD) simulations were carried out to investigate the behavior of a nanoscale tip sliding on graphene samples with different strains. In the simulations, a rigid silicon tip was used to slide on 2 types of surfaces, i.e., a graphene sheet supported on an amorphous Si (a-Si) substrate, and freely suspended graphene sheets with different magnitudes of biaxial strain (see Materials and Methods for more details). As shown in SI Appendix, Fig. S6, both the atomic-scale stick–slip and the 2-stage friction strengthening effect are successfully reproduced in the simulations. The 2-stage friction strengthening effect has been discussed in the earlier study (19). The possible mechanisms are briefly outlined in the following. When graphene is suspended, it can deform freely and the configuration of the tip–graphene contact interface can reach the steady state immediately upon contact. Therefore, no obvious evolution effect can be observed. However, when graphene is supported on a substrate, the situation is quite different. Upon contact with the tip, the graphene sheet would be sandwiched between the tip and the substrate. Due to the geometrical constrain, graphene cannot deform as freely and quickly as in the suspended case. As reported in the earlier study (19), when the tip slides forward, the contact zone gradually evolves by progressively collecting the looseness from the surrounding, which results in an enhanced interface pinning capability. In addition to the evolution of the pinning capability, the coordination of the interfacial pinning forces also evolves gradually each time when the tip reverses its slide direction, resulting in the repeated evolution effect of the lateral force traces (more discussions can be found in SI Appendix, Fig. S7). Besides the evolution effect, our calculation results show a consistent trend that the stick-and-slip asymmetry and friction force decreases with increasing tensile strain of graphene. For example, the peak lateral force during the steady state is reduced from 6.55 nN for loosely supported graphene, to 3.07 nN for suspended graphene when gently compressed (rippled), and eventually to 0.55 nN when slightly stretched, as shown in Fig. 3A (more discussions can be found in SI Appendix, Fig. S8). By counting the number of atoms that are closely interacting (within a cutoff distance of 0.4 nm), we found that the changes in the tip–graphene contact area (see SI Appendix, Fig. S9) could not account for the friction variation under different strains.

As previous work has shown that the contact quality of the sliding interface can significantly affect the frictional behavior of 2D materials (19), we analyzed the distribution of the atomic-scale interaction forces \( f_i \) along the interface. The typical distributions of the interaction forces acting on the tip when the lateral force reaches the local peak values are shown in Fig. 3B ( Insets) for the supported graphene and the suspended graphene. Because of the incommensurate contact between the tip and graphene, \( f_i \) shows a random distribution. As the graphene changes from the compressed (rippled) to the stretched state, the
friction were calculated by performing a linear fit of the mean friction vs. normal force curves in repeated measurements under the same normal load. The kurtosis parameter is a measure of the tailedness of the interfacial force distribution. A larger kurtosis value implies that there are more interfacial force distributions given by a distribution. A larger kurtosis value implies that there are more interfacial forces having higher magnitudes. As shown in Fig. 3B, the kurtosis parameter decreases monotonically when the strain varies from −0.4 to 0.3%, which means that there are less interfacial atoms being strongly pinned when graphene is stretched. This variation is closely related to the deformability of the graphene sheet as illustrated in SI Appendix, Fig. S10. This behavior is understandable because applying a strain in graphene would change the out-of-plane flexibility, thereby affecting the contact quality (19). As illustrated in Fig. 3C and D, a compressive strain renders graphene a more relaxed configuration and results in a more intimate contact, whereas a tensile strain constrains the flexibility of graphene producing a less-pinned interface. The dominating contribution of the deformability of graphene to friction was further confirmed by artificially altering the stiffness of graphene in MD simulations (see more details in SI Appendix, Fig. S11).

The atomistic simulations suggest that the friction tuning is achieved by altering the flexibility of graphene through applying in-plane strain, which should be a completely reversible process. To confirm this hypothesis, we carried out friction measurements while applying a cyclic load on graphene. Fig. 4A shows the mean friction versus normal force curves acquired on graphene while the applied strain alternates between 0.0 and 0.1%. We can clearly see that the effect of strain on friction is indeed reversible and the friction can be fully recovered when the strain is released (see more details in SI Appendix, Fig. S12). We also calculated the COF during each cycle and plotted them in Fig. 4B. As the strain changes cyclically, the COF varies consistently, demonstrating the high reliability of this friction-tuning method.

In conclusion, friction is one of the most common yet most complicated phenomena due to its intricate origins and multiscale nature. Tuning friction in a dynamic, reversible, and controllable way has long been a great challenge in the mechanics...
and materials community. By adjusting the in-plane strain of graphene, we demonstrate that the surface friction of graphene can be well regulated by altering the atomic-scale contact quality. Our work gives the very example, where the atomic-scale interfacial interactions can be directly regulated via macroscopic operations. The proposed friction-tuning strategy is expected to be applicable for other sliding interfaces with configurationally flexible components.

**Materials and Methods**

**Sample Preparation.** The silicon wafer covered with a 300-nm-thick SiO₂ layer was purchased from Silicon Valley Microelectronics. The round-hole array was fabricated by photolithography and reactive ion etching on the Si/SiO₂ wafer. The depth and the diameter of the holes were 300 nm and 5 μm, respectively. The graphene was prepared by mechanical exfoliation and then transferred to the prepatterned Si/SiO₂ substrate. For graphene straining, we first placed the graphene samples in an autoclave for 2 d to get equilibrated with a preset high pressure (−0.18 MPa). Then the graphene samples were taken out in ambient conditions and the graphene sheets would buckle, resulting in a tensile strain due to the pressure difference. After the friction measurements, we placed the sample in a vacuum chamber with a low pressure (−0.01 MPa). By controlling the storage time in the vacuum chamber, we could reduce the pressure difference across the graphene layer and release the strain. To get cyclic straining, we repeated the above steps. The thickness of graphene layers was identified by Raman spectroscopy (Renishaw system, laser wavelength of 514 nm). The laser intensity was set below 0.1 mW to minimize the influence of laser heating and the spatial resolution was 1 μm. The Raman spectra were fitted with the Lorentzian function to determine the peak positions.

**Sample Measurements.** The AFM from Asylum Research (model: Cypher) was employed to perform the friction, topography, and adhesion measurements in ambient conditions (20–25 °C, relative humidity 20–30%). For the friction and topography measurements, silicon nitride probes (Budget Sensors, SiNi) were used and the normal and lateral force constants were calibrated by thermal noise method and a diamagnetic lateral force calibrator, respectively. The mean frictions in Fig. 1C and SI Appendix, Fig. S3 were determined by calculating the half difference of the trace and retrace lateral force signals, which were obtained in a square region of 50 nm × 50 nm under a sliding velocity of 500 nm s⁻¹. Silicon probes (MikroMasch, CSC37/Al BS) were used to perform adhesion measurements shown in Fig. 2B and the pull-off force from the force-displacement curves was reported as the adhesion force.

**MD Simulation.** The atomistic simulations were carried out to study the stick-slip friction of a rigid silicon tip sliding on 2 graphene systems, i.e., monolayer graphene supported on an amorphous silicon (a-Si) substrate and suspended monolayer graphene with different amounts of preapplied bi-axial strain. The C-C interaction of graphene and the Si-Si interaction in the substrate and the tip were described by Tersoff and Stillinger-Weber potentials, respectively. The graphene–tip and graphene–substrate interactions were modeled by typical 6–12 LJ potentials. The LJ parameters were set (ε = 0.008 eV, σ = 0.3 nm for graphene–substrate interaction and ε = 0.005 eV, σ = 0.3 nm for graphene–tip interaction). To match the work of adhesion from simulations with the experimental values. The lateral dimension of the monolayer graphene sheet is 38.5 nm × 38.6 nm. The a-Si substrate was prepared by quenching liquid silicon under a rate of 10¹³ K/s and had a lateral dimension of 43.4 nm × 43.4 nm and a thickness of around 3.0 nm. The bottom atoms of the substrate were fixed. The rigid <001>–oriented crystalline silicon tip was constructed with a bowl-like shape (radius of 16.3 nm). The graphene atoms at the edges of graphene sheet were fixed rigidly to constrain the overall translational movement. To incorporate the compliance of the AFM cantilever in experiments, we coupled a harmonic spring in the x direction to pull the tip. The constant of the lateral spring was chosen to be 11.2 N/m for all simulations, comparable to the typical experimental value. Since the main focus of this work is about tuning friction (or energy corruagion) via in-plane straining, we only considered the quasi-static behavior of the system. In the simulation, we gradually changed the tip and substrate relative displacement at each stage and distance to minimize the influence of loading speed so that quasi static behavior could be obtained. The stick–slip motion was performed under a quasistatic loading. Specifically, the lateral spring was displaced along the x direction with a prescribed distance (0.03 nm) in each loading step and then relaxed for 20 ps. To minimize the large oscillation of tip and facilitate the damping during relaxation, a viscous force was applied to the spring as f = −v Δx (v = 1.6 × 10⁻⁹ N/(m/s)) is the drag coefficient and v is the velocity of the tip. Attention was paid to make sure the impact the viscous damping force was minimal at the final moment of relaxation. The simulation was performed at 300 K using a Nosé–Hoover thermostat. We used LAMMPS code for calculations and AtomEye for visualization.

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