Disorder-induced magnetoresistance (MR) effect is quadratic at low perpendicular magnetic fields and linear at high fields. This effect is technologically appealing, especially in 2D materials such as graphene, since it offers potential applications in magnetic sensors with nanoscale spatial resolution. However, it is a great challenge to realize a graphene magnetic sensor based on this effect because of the difficulty in controlling the spatial distribution of disorder and enhancing the MR sensitivity in the single-layer regime. Here, a room-temperature colossal MR of up to 5000% at 9 T is reported in terraced single-layer graphene. By laminating single-layer graphene on a terraced substrate, such as TiO2-terminated SrTiO3, a universal one order of magnitude enhancement in the MR compared to conventional single-layer graphene devices is demonstrated. Strikingly, a colossal MR of >1000% is also achieved in the terraced graphene even at a high carrier density of \( \approx 10^{12} \text{ cm}^{-2} \). Systematic studies of the MR of single-layer graphene on various oxide- and non-oxide-based terraced surfaces demonstrate that the terraced structure is the dominant factor driving the MR enhancement. The results open a new route for tailoring the physical property of 2D materials by engineering the strain through a terraced substrate.

Hybrid 2D material/complex oxide interface, combining the layered 2D materials and complex oxides, provides various opportunities to study the intriguing physics and develop multifunctional devices, due to the rich properties of both 2D materials and complex oxides, such as 2D electron gas, ferromagnetism, ferroelectricity, and superconductivity.\(^{1,2}\) The interface coupling between 2D materials and complex oxides will alter the lattice structure and induce the entanglement of the charge, spin and orbital degrees of freedom in 2D materials. The interface is crucial to improve the functionality of 2D material-based devices.

Magnetoresistance (MR), the change in the electrical resistance under an external magnetic field, not only provides an efficient tool for probing the fundamental electronic properties in materials, but also results in advanced technological applications.\(^{9}\) However, intrinsic graphene does not exhibit a MR effect.\(^{10,11}\) Nevertheless, graphene films can display strong MR by harnessing disorder. According to the classical

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disorder-induced MR effect, the origin of MR in graphene can be attributed to the fluctuation in carrier density and mobility.[12,13] Various efforts have been devoted to engineering disorder in single-layer graphene, such as by decorating with gold nanoparticles,[14] by fluorination or nitrogen doping,[15,16] or by stacking on a lattice-mismatched substrate.[17] However, the MR of single-layer graphene has yielded limited success thus far at room temperature, typically ranging between 60% and 775% at 9 T (Table S1, Supporting Information).[12–23] The multilayer graphene can usually exhibit a large MR due to its interlayer effect. For example, six-layer graphene on hexagonal boron nitride (BN) has been reported to show large room-temperature MR of ≈2000% at 9 T.[24] However, the precise control of the layer number in multilayer graphene produced by the mass production techniques, such as roll-to-roll (R2R) or batch-to-batch (B2B) methods, remains challenging.[25] Moreover, as previously reported,[12–24] the MR of graphene will rapidly decay to below 200% at a higher carrier density of 10^{12} cm^{-2}, which means unintentional doping will lead to the loss of MR sensitivity. The fast batch production of single-layer graphene films, on the other hand, raises the prospect of fabricating a robust graphene magnetic sensor based on a single-layer.[26,27]

Here, we realized a room-temperature colossal MR of up to 5000% at 9 T, by designing terraced single-layer graphene. We found that the colossal MR of the terraced graphene does not rely on the surface termination of the substrate. Both oxide-based (TiO_2, AlO_2, or FeO_2) and nonoxide-based (AlN) surface terminations produced colossal MR effects consistently above 1000% even at a high carrier density of ≈10^{12} cm^{-2}. Inspired by this finding, we artificially fabricated terraces and steps on a commonly used BN substrate and found that the room-temperature MR of graphene on the terraced BN can also be enhanced to more than 1000%. Combined scanning tunneling microscopy (STM) and density functional theory (DFT) calculation revealed that the topographic corrugations and inhomogeneous charge puddles both result in disorder in the terraced graphene, which significantly enhances the scattering when applying a magnetic field. Finally, we analyzed and fit our MR and Hall data using the self-consistent effective medium theory.

The terraced single-layer graphene is formed by stacking single-layer graphene on an atomically terraced substrate (Figure 1a). After the lamination of the graphene on the terraced substrate (Figure S1, Supporting Information), the van der Waals (vdW) interaction between the graphene layer and the substrate is expected to be enhanced due to the elastic deformation of graphene, inducing strain both in the terraces and steps.[28] To realize the terraced single-layer graphene, a perovskite-oxide single crystal, STO, is utilized as a prototypical substrate for various following reasons. First, atomically flat terraces and steps can be robustly reproduced on the STO surface by selectively etching the SrO layer, leading to a TiO_2 terminated surface with a unit-cell (≈0.4 nm) step height (see the Experimental Section).[29,30] Second, STO can induce an inhomogeneous topographical corrugation in graphene due to a large lattice mismatch between graphene and STO, which also contributes to the disorder in graphene.[13] Third, STO has a large dielectric constant at room temperature (κ ≈ 200–300) with a wide bandgap of 3.2 eV, providing a strong dielectric screening of charged impurity scattering to ensure the high mobility in graphene is maintained.[14,32] Finally, the possibility of (epitaxially) growing various other materials on STO allows us to examine the influence of surface terminations on the MR.[33]

To fabricate terraced single-layer graphene, graphene flakes were first exfoliated onto a poly(methyl methacrylate)/poly(methyl glutarimide) (PMMA/PMGI) coated silicon substrate. After identification by Raman spectroscopy, single-layer graphene was transferred onto the terraced STO substrate using the dry-transfer technique (see the Experimental Section). After the lamination of graphene on the terraced STO, the surface of graphene showed similar terrace and sharp-step features as those of STO (Figure 1b). To estimate the localized strain induced by the atomic steps, we probes the surface of graphene on STO (G/STO) and the bare STO by line profile scanning using atomic force microscopy (AFM) (Figure 1c). Similar to the bare STO surface, the graphene surface also showed atomically flat terraces with sharp steps of around 0.4 nm, indicating that the graphene layer has adapted a terraced structure. Because of the elastic deformation, a tensile strain of up to 0.2% can be induced in the steps of the terraced graphene, which can be confirmed by analyzing the Raman 2D mode of single-layer graphene (Figure S2, Supporting Information).[34,35] Under elastic deformation, the applied stress σ is proportional to the strain ε, σ = Ye, where Y = 1 × 10^{10} Pa denotes Young’s modulus of the PMMA film,[34] and thus the PMMA/Terraced graphene is expected to receive applied stress of ≈2 × 10^{6} Pa, enhancing the vdW interaction between the terraced graphene and STO. As shown in the height histogram of G/STO (Figure S3, Supporting Information), the average height difference between graphene and STO is only 0.44 nm, indicating single-layer graphene with a narrow vdW gap of ≈1 Å.[36] Subtracting the Raman spectra of G/STO from the bare STO leads to a single Lorentzian peak at 2680 cm^{-1} (2D mode), which is 2.2 times of the sharp peak at 1588 cm^{-1} (G-mode), further confirming the single layer of the terraced graphene (Figure 1d).[26]

To explore the MR effect in the terraced single-layer graphene, we first investigated the MR of currents traveling either parallel or perpendicular to the steps using standard Hall bar devices (see the Experimental Section and Figure S4 in the Supporting Information). Since the MR perpendicular to the steps (1050%) is slightly larger than the MR parallel to the steps (800%), we focus our study on the case of current perpendicular to the steps. Figure 1e shows the top-gate dependence of the longitudinal resistance (R_{xx}) at 300 K. At zero magnetic field, a maximum resistance can be observed at 0.5 V which corresponds to the charge neutrality point (CNP) and indicates p-doping of the terraced graphene.[17,24] Applying a magnetic field of 9 T perpendicular to the sample surface results in a significant increase of R_{xx} for all ranges of the top gate voltage. To quantify the MR effect, we follow the conventional definition that MR = 100% × (R_{xx}(B) − R_{xx}(0))/R_{xx}(0), where R_{xx}(B) and R_{xx}(0) are the resistance at B and zero fields, respectively. The observed MR typically peaks at the CNP and gradually decreases while increasing or decreasing the gate voltage away from the CNP, as shown in the blue curve of Figure 1e. This gate tunable MR feature is consistent with the previous reports[14,17,24] and can be qualitatively understood by the two-fluid model.[17] Figure 1f shows R_{xx} and MR as a function of a magnetic field (B) near the CNP. As the magnetic field is increased, R_{xx} initially...
shows a quadratic increase in low fields, subsequently turns into a linear increase at moderate fields, and finally saturates at 9 T (Figure S5, Supporting Information). The crossover from the quadratic to linear MR behavior is the hallmark of the classical disorder-induced MR. Remarkably, the MR of the terraced single-layer graphene at the CNP is found to be as high as 5000% at 9 T, which is one order of magnitude higher than that reported on previous single-layer graphene devices at the same conditions (Table S1, Supporting Information).

To confirm that the colossal MR is due to the formation of a terraced structure in graphene induced by the surface topography of the substrate, graphene was laminated on either nonterraced STO surface or BN-buffered terraced STO. First, placing graphene on the as-received STO (without terraces and steps) resulted in a low MR of \( \approx 170\% \) at 9 T and 300 K (Figure S6, Supporting Information), similar to the MR of graphene on SiO\(_2\). A clear quantum oscillation was observed at low temperatures with nearly zero MR at low fields, resembling graphene on a flat surface and emphasizing the absence of the substrate corrugation effect. Second, introducing a thick BN layer of 20–30 nm as a buffer between graphene and the terraced STO, thus the steps of the terraced STO can be completely screened by BN, led to a low MR of \( \approx 215\% \) (Figure S7, Supporting Information), in agreement with the previous report of single-layer graphene on BN. These results demonstrate that without the steps and terraces, the composition (STO, SiO\(_2\), and BN) of the surface does not contribute to the enhancement of the MR. Only the surface topography, in particular the atomic steps in STO that enhances the MR in single-layer graphene.

Next, we study how the surface termination influences the colossal MR of the terraced graphene by investigating a range of oxide surface terminations including TiO\(_2\)-terminated BaTiO\(_3\), AlO\(_2\)-terminated LaAlO\(_3\) and FeO\(_2\)-terminated LaFeO\(_3\) (Figure 2a–c). To verify whether an oxide termination is necessary for obtaining the colossal MR, we also study a nonoxide terminated surface AlN (Figure 2d). All of the surface terminations were prepared by growing the respective thin film...
using pulsed laser deposition (PLD) on a terraced STO substrate.\[33\] Before the thin film growth, the presence of the terraces and steps on the STO substrates was verified by atomic force microscopy (AFM). Six-unit cells of the oxide films were then grown on the STO substrate and monitored by reflection high-energy electron diffraction (RHEED) (Figure 2e). Both the periodic RHEED oscillation and streaky RHEED pattern before and after growth demonstrated the layer-by-layer growth of high-quality thin films. Single-layer graphene was subsequently transferred onto the freshly prepared surface. Strikingly, all surfaces resulted in a colossal MR >1200% for oxide-based terminations (TiO2, AlO2, or FeO2) at a relatively high carrier density of $n > 1.2 \times 10^{12}$ cm$^{-2}$ (Figures S8–10) and ≈1060% for nonoxide based termination (AlN) at the carrier density of $1.8 \times 10^{12}$ cm$^{-2}$ (Figure S11, Supporting Information). For comparison, previous reports of single-layer graphene prepared on SiC, SiO2, BN, and BP showed a very low MR of around 150% at high-level doping of $10^{12}$ cm$^{-2}$.[14–24] These results suggest that the MR of the terraced single-layer graphene is not dependent on the surface terminations but the surface terraces and steps of the substrate. Inspired by this finding, we artificially fabricated the terraces and steps on a commonly used BN substrate and found that the room-temperature MR of graphene on the terraced BN can also be enhanced to 1100% at 9 and 300 K, while it is only 160% for the graphene on flat BN (Figure S12, Supporting Information), indicating the crucial role of the terraces and steps of BN in enhancing the magnitude of the MR.

To better understand the MR characteristics of the terraced single-layer graphene, we studied the MR changes with gate voltage, temperature and magnetic field direction in the G/STO system (Figure 3). Figure 3b shows a gate tunable colossal MR in the terraced single-layer graphene. The MR is maximum close to the CNP and gradually decreases with decreasing gate voltage away from the CNP, indicating the intrinsic character of the terraced graphene is the source of the colossal MR.[14] While the MR shows saturation when lowering the carrier density, there is no sign of saturation with the increase in the carrier density. The observation of nonlinear Hall behavior (Figure 3c,f) can be associated with the coexistence of two densities of carriers.[24] Figure 3e shows temperature-dependent MR near the CNP. At 2.5 K, apart from the Shubnikov–de Haas oscillations, aperiodic oscillations with a much smaller amplitude of around 0.03 e$^2$ h$^{-1}$ can also be seen (Figure S13e, Supporting Information), consistent with the criteria that suggest the presence of universal conductance fluctuation (UCF) in our terraced graphene samples.[40,41] With the increase in temperature, the oscillations are rapidly suppressed and completely vanish due to the thermal broadening of Landau levels. Interestingly, above
100 K, the slope of the Hall resistance changes sign from negative to positive (Figure 3f), indicating a change in the dominant carriers from electrons at low temperature to holes at high temperature (Figure S13, Supporting Information), as previously also observed in the graphene/LaAlO$_3$ (LAO)/STO system. The temperature-dependent resistance behavior near the CNP (Figure S13c, Supporting Information) indicates the disorder-dominated transport in terraced graphene. Another evidence that the terraced graphene is consistent with the classical MR picture is the angle dependence of MR (Figure 3d). $R_{xx}$ reaches a maximum value when the magnetic field is normal to the plane (0° and 180°) and a minimum value when the magnetic field is parallel to the plane (90° and 270°), consistent with the classical Lorentz force causing the deflection of the trajectory of the charged particles.

Finally, to study the localized disorder of graphene on STO at the nanoscale, we conducted scanning tunneling microscopy (STM) and noncontact atomic force microscopy (nc-AFM) studies of graphene on STO. STM topography imaging shows a random corrugation with bumps and depression regions (Figure 4a). The atomic-scale image (inset) shows the graphene honeycomb lattice with lattice distortion. The derived surface strain can be as large as 1.7%, as calculated from the measured displacement in the graphene lattices (Figure S14, Supporting Information). A comparison between nc-AFM and STM topography over the same area yields apparent correlation (Figure 4b) between topographic corrugation and electronic fluctuation, indicating that the strain-induced topographic corrugation is the dominant contributing factor for the observed surface fluctuation. Moreover, the shifting of the Dirac points, nearly 30 meV, at bump and depression area indicates that there are inhomogeneous charge puddles on the surface (Figure 4c), which can be further verified by the $dI/dV$ mapping near the Dirac point.[44] The $dI/dV$ maps at a bias below and above Dirac point showing a nearly inversed spatial density of states (DOS) distribution indicate the different shifting of the Dirac point at different spatial areas with inhomogeneous charge puddles (Figure S15, Supporting Information). As a result, graphene on STO contains both disorders of topographic corrugations and inhomogeneous charge puddles at the terrace, which significantly enhance the electron scattering in a magnetic field.

To understand the origin of the localized strain in the terraced graphene, we performed DFT calculations to gain insights into the interfacial interaction between graphene and STO (see the Experimental Section). Due to a large mismatch of the lattice constants between graphene and STO, the graphene may form incommensurate superlattice structures on STO, in which the different regions of graphene experience variable strains from the STO substrate. Therefore, in the simulation, we considered two different manually strained graphene supercells on the STO substrate (see the details in the Experimental Section). As Figure 4d,e shows, the optimized structure shows that graphene on STO with the larger compressive strain (≈6.9%) becomes warped. The highly strained graphene results in a pronounced charge inhomogeneity in the graphene (Figure 4e). However, the electronic structure of graphene does not change significantly (Figure 4f) since the Dirac cone of graphene can still be seen in the band structure,[45] consistent with the $dI/dV$ curve measured by STM. For graphene on the BN substrate, the optimized structure of graphene is flat and the charge density redistribution is uniform (Figure S16, Supporting Information) due to the matched lattice constants between graphene and BN. Moreover, the interfacial interaction of G/STO is stronger than that of G/BN and G/BP, as evidenced by the larger adsorption
energy, a shorter interfacial spacing, and more pronounced charge redistribution (Figure S17, Supporting Information). The stronger interaction and shorter spacing between graphene and STO facilitate the effective coupling between graphene and STO, leading to stronger scattering due to charge puddles in graphene and phonon scattering from the substrate. On the other hand, the preserved Dirac cone of graphene on the STO substrate indicates that the carrier mobility of strained graphene does not degrade significantly. All these may contribute to the observed colossal MR in the terraced graphene.

Various theoretical models have been proposed to understand the disorder-induced MR effect. The two-fluid model assumes the coexistence of electrons and holes at the CNP that can explain the sharp MR peak at the CNP but with a saturating MR feature in high fields. The model also predicts that the MR approaches zero far away from the CNP, which is consistent with our observation. The other two models that have successfully described the disorder-induced MR are the random resistor network model and the self-consistent effective medium theory, which have been demonstrated to belong to the same universality class. The effective medium theory considers the presence of electron–hole puddles and the strong distortion of current lines at the boundaries between n and p-type puddles under a magnetic field. Based on the effective medium theory, we successfully fit the MR data (Figure 3b) and the Hall data (Figure 3c) near the CNP. Here, different parameters are used to fit the MR and Hall data, and the discrepancy may be attributed to the various assumptions used in this model (see Figure S18 in the Supporting Information and its description).

The band structures of 2D semiconductors like transition metal dichalcogenides are usually sensitive to the strain. For example, the bandgap of MoS2 can be modulated by the tensile strain, leading to excellent electrical and optical performance. Our STM and DFT calculations (Figure 4c,f) on the contrary show that the electronic structure of terraced graphene does not change significantly with the application of strain, resulting in a robust carrier mobility. In a disordered system, this high mobility is expected to contribute to the large MR. Note that our MR in terraced graphene based on standard Hall bar structure is the
physical MR, not from the geometrical effect, since either the physical or geometrical contribution may dominate the observed MR. The MR can be further enhanced by the geometrical effect, like extraordinary magnetoresistance (EMR). Moreover, not only can the strain enhance the MR effect in terraced graphene, but it may also lead to the pseudomagnetic field (PMF). This is because a nonuniform strain can deform the corners of the graphene Brillouin zone, shifting the Dirac cones at K and K' points in the opposite directions, creating PMF. The strain-induced large MR and PMF effects have been previously demonstrated in G/BP system. We believe our terraced graphene systems, apart from the MR effect, will also exhibit the PMF effect which would be an interesting future study. Recent electrical transport and STM studies of PMF in terraced graphene indicate that our proposal is plausible.

In conclusion, we designed and demonstrated a viable way to artificially engineer disorder in single-layer graphene by laminating it on a terraced substrate. This results in a strained and terrace-like single graphene layer that exhibits a colossal MR of one order of magnitude larger than that of conventional flat graphene at room temperature. The MR of the terraced graphene is gate tunable, nearly temperature independent, and magnetic field angle-dependent following the classic disorder-induced MR effect. Unlike the enhanced MR effect previously obtained by chemical doping or surface decoration of the graphene layer which modifies the electronic band structure of graphene, the disorder-induced MR by substrate terraces is highly reliable, reproducible, and nondestructive. The intrinsic property of graphene is well maintained. Most importantly, the MR sensitivity remains robust even at a high-doping level of $10^{12}$ cm$^{-2}$. Our methods open a new route for tailoring the physical property of 2D materials, especially for those strain-sensitive 2D semiconductors such as MoS$_2$, MoTe$_2$ or 2D phene. Brillouin zone, shifting the Dirac cones at K and K' because a nonuniform strain can deform the corners of the graphene Brillouin zone, shifting the Dirac cones at K and K'.

**Experimental Section**

**HF Treatment of STO (001):** To obtain the atomically flat terraces and steps in STO, the as-received (001)-oriented STO single crystalline substrate (CrysTec GmbH) was first ultrasonically soaked in analytical grade and demineralized water for 10 min to form Sr-hydroxide complex, followed by a 30 s dipping in standard and commercially available buffered HF solution. The chemical reactions are: SrTiO$_3$ + H$_2$O = Sr(OH)$_2$ + TiO$_2$, Sr(OH)$_2$ + 2 HF = SrF$_2$ + 2H$_2$O. During the process, the SrO layer will be chemically selectively dissolved by HF, ensuring that the surface is purely TiO$_2$-terminated with atomically flat terraces and steps. To remove the remnants of the previous treatments and facilitate recrystallization, a final annealing step was performed at 950 °C for 2 h. The surface topography of the terraced STO was characterized by atomic force microscopy (AFM) (Bruker Icon3, Bruker Corporation, USA) with a repetition rate of 2 Hz. BTO, LAO, and LFO thin films were grown under 30 s on SrTiO$_3$ in 0.8 J cm$^{-2}$ and a repetition rate of 10 Hz and subsequently annealed at 550 °C for 2 h to remove oxygen vacancy in STO.

**Device Fabrication and Characterization:** To fabricate a terraced single-layer graphene device, natural graphite (NGS, Germany) was first mechanically exfoliated on a PMGI/PMMA-coated silicon substrate. After identifying the single-layer graphene by Raman spectroscopy (Renishaw), the PMGI layer was slowly dissolved by the MF319 solution, and subsequently, the graphene/PMMA bilayer was cleaned thoroughly in DI water. The graphene/PMMA bilayer was then transferred onto the terraced substrate using the dry-transfer technique. Finally, the PMMA layer was removed by acetone and terraced single-layer graphene was finally obtained. To fabricate a Hall bar channel, either parallel or perpendicular to the steps of the substrate, a Au marker (10 nm thick) was deposited around the graphene flake and then annealed at 350 °C for 2 h under H$_2$/Ar gas to remove the PMMA residue. The angle between the steps and horizontal lines of the Au marker can be clearly identified by AFM. Based on the measured angle, the C/Au (2/60 nm) electrodes can be exactly designed and deposited both parallel and perpendicular to the steps using Lesker Thermal Evaporator. Finally, O$_2$ plasma was used to obtain 8 μm × 2 μm graphene stripes.

**Electrical Transport Measurement:** The electrical transport measurements were carried out in a physical property measurement system (PPMS) interfaced with a source meter (Model 2400, Keithley Inc.) and a multimeter (Model 2002, Keithley Inc.). A direct current of 1 μA was applied through the sample to measure the longitudinal resistance in a magnetic field (perpendicular to the sample plane) with the magnitude varying from ~9 to 9 T. Before the measurement, the samples were annealed for 2 h at 350 K in H$_2$/Ar atmosphere to remove the PMMA residual and any adsorbed water vapor. To apply top-gate bias, the source terminal was connected to the top gate electrode and the leakage current through the BN layer was monitored. At low magnetic fields between 0 and 1 T, the linear slope $s = -\frac{dR}{ds}$ determines the carrier density $n = 1 se$. The Hall mobility is calculated through the expression $\mu = \frac{1}{neR_H}$.

**STM/nc-AFM Imaging:** STM/nc-AFM measurements were carried out in a LT-Omicron STM system (base pressure $1 \times 10^{-10}$ mbar). C/STO device was degassed to 600 K to get rid of surface contamination and then transferred into the STM head to cool down to 77 K for STM/nc-AFM scanning. qPlus sensor with electrochemically etched tungsten tips was used. Bias voltages were applied to the sample for STM and STS measurements. STS was obtained by the built-in lock-in amplifier in the Nanonis controller, with which a 20 mV (rms), 963 Hz sinusoidal modulation was superimposed on the sample bias. For nc-AFM imaging, the constant height mode with an oscillation amplitude of about 2 Å was used to record the frequency shift ($\delta f$). All the STM/nc-AFM images were processed using WSXM software.

**Theoretical Calculations:** All calculations were carried out using density-functional theory (DFT) based on Vienna ab initio simulation package (VASP5.4.18) with the Perdew–Burke–Ernzerhof (PBE) approximation for the exchange-correlation functional and projector-augmented wave (PAW) potential for the electron–ion interaction. The cut-off energy for the plane-wave expansion was set to 500 eV, and the vacuum layer with a thickness larger than 15 Å was applied in all the calculations. An effective Hubbard $U$ ($U = 4.0$) was applied to orbital electrons of Ti. For all the hybrid structures, van der Waal interaction was included using the DFT-D3 method. The electronic conduction $T$ was set to $1.0 \times 10^{-3}$ eV, and the force on each atom is optimized smaller than 0.01 eV Å$^{-1}$.

For the interface models of graphene on the STO substrate, two different graphene supercells were considered on the STO substrates. One was to place $(\sqrt{3} \times 5)$ graphene supercell on the (1 1 3) STO substrate, in which about 6.9% and 3.2% compressive strain were applied along the armchair and zigzag edge of graphene, respectively (large strain, L-C/STO). Another was based on $(4 \sqrt{3} \times 7)$ graphene supercell on the $(3 \sqrt{2} \times 3 \sqrt{2})$ STO, where the armchair and zigzag edge...
of graphene were compressed by about 1.3% and 2.3%, respectively (small strain, 5-C/STO). For the interface model of graphene on a h-BN substrate, graphene was stretched by about 1.3%. Besides, the interface structure of graphene on the black phosphorus (BP) substrate (G/BP) was constructed by placing strained $4 \times \sqrt{3}$ graphene supercell on the bilayer (3 \times 1) BP supercell, with a tensile strain of about 1.1% and 1.6% applied on the zigzag and armchair edge of graphene, respectively. T-centered $12 \times 12 \times 1$, $10 \times 10 \times 10$, $8 \times 4 \times 1$, $2 \times 2 \times 1$, and $4 \times 12 \times 1$ k-point meshes were used for sampling the Brillouin zones of graphene (graphene/h-BN), STO bulk, L-G/STO, S-G/STO, and G/BP, respectively.

The adsorption energy for graphene on STO, h-BN, and BP was estimated by $\Delta E_{ad} = E_G + E_X - E_{G,X}$, where $E_G$ is the total energy of the hybrid structures for graphene on STO, BN, or BP; $E_X$ is the total energy of isolated graphene, and $E_{G,X}$ is the total energy of isolated STO, BP, or BP layers, respectively. The larger $\Delta E_{ad}$ indicates stronger interfacial interaction between graphene and the substrate. The charge density redistribution for graphene on the substrates can be seen from the differential charge density difference as defined by $\Delta \rho = \rho_{G,X} - (\rho_G + \rho_X)$, in which $\rho_{G,X}$ is the charge density of the hybrid structures, and $\rho_G$ and $\rho_X$ are the charge density of isolated graphene and the substrate (STO, h-BN, or BP), respectively.

Data Availability
The data that support the plots within this paper and other findings of this study are available from the corresponding authors upon reasonable request.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

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