Robust quantum spin Hall effect in graphene induced by dielectric screening

Louis Veyrat,1 Corentin Déprez,1 Alexis Coissard,1 Xiaoxi Li,2,3,4 Frédéric Gay,1 Kenji Watanabe,5 Takashi Taniguchi,5 Zheng Han,2,3,4 Benjamin A. Piot,6 Hermann Sellier,1 and Benjamin Sacépé1

1 Univ. Grenoble Alpes, CNRS, Grenoble INP, Institut Néel, 38000 Grenoble, France
2 Shenyang National Laboratory for Materials Science, Institute of Metal Research, Chinese Academy of Sciences, Shenyang 110016, P. R. China
3 School of Material Science and Engineering, University of Science and Technology of China, Anhui 230026, P. R. China
4 State Key Laboratory of Quantum Optics and Quantum Optics Devices, Institute of Opto-Electronics, Shanxi University, Taiyuan 030006, P. R. China
5 National Institute for Materials Science, 1-1 Namiki, Tsukuba 306-0044, Japan
6 Laboratoire National des Champs Magnétiques Intenses, LNCMI-CNRS-UGA-UPS-INS-A-EMFL, F-38042 Grenoble, France

The quantum spin Hall (QSH) effect has the remarkable property of alloying an insulating bulk with conducting helical edge states1,2. Such a topological phase holds many promises for quantum transport, spintronics3 and topological quantum computation4. Since its discovery in HgTe/CdTe quantum wells5,6, the search for easy-to-produce, new QSH systems combining a large bandgap for operation at high temperature with long length scale helical edge transport, remains tremendously active and challenging. In graphene, the zero-energy Landau level (zLL) emerging at charge neutrality under perpendicular magnetic field has been early put forth as an alternative candidate7,8. The lifting of the zLL spin and valley degeneracies into broken-symmetry states was expected to result in an unusual spin-polarized, ferromagnetic ground state9,12,16,17, recently termed quantum Hall topological insulator12, harboring two counter-propagating, helical edge channels. However, most experiments in clean graphene devices show an otherwise insulating state at charge neutrality with gapped bulk and edge excitations, which is accounted for by a strong renormalization of the symmetry-breaking interaction terms by Coulomb interaction11. Here we show that the ferromagnetic quantum Hall ground state can be induced by screening the long-range Coulomb interaction in graphene with the use of SrTiO3 as a high-k dielectric substrate, yielding a robust and gate-tunable QSH effect. This state features non-local transport properties characteristic of current-carrying helical edge channels, which emerge under a magnetic field as low as 1 T and persist up to a temperature of 110 K. At high magnetic field, a transition towards insulation develops, indicating a gap opening for the edge excitations. Our work demonstrates how suitable substrate screening can tune an interaction-driven topological quantum phase transition between trivial and topological ground states12. The resulting quantum Hall topological insulator in graphene on high-k dielectric devices provides a new and versatile platform hosting helical edge channels, which can be easily coupled to superconducting electrodes13, magnetic materials14 or local gates15 for investigating spin-transport13,14,16 and hybrid superconducting circuits based on Majorana17,19 or parafermions20,21 non-Abelian states.

There is a variety of topological phases that are classified by their dimensionality, symmetries and topological invariants1,2,2,3. They all share the remarkable property that the topological bulk bandgap closes at every interfaces with vacuum or a trivial insulator, forming conductive edge states with peculiar transport and spin properties. The quantum Hall effect that arises in two-dimensional electron systems subjected to a perpendicular magnetic field, B, stands out as a paradigmatic example characterized by a Z invariant—the Chern number—which quantizes the Hall conductivity and counts the number of chiral, one-dimensional edge channels. The singular aspect of quantum Hall systems compared to quantum wells is the spontaneous breaking of the SU(4) symmetry splinters the Landau levels into quartets of broken-symmetry states that are polarized in one, or a combination of the spin and valley (pseudospin) degrees of freedom33–35.

Central to this phenomenon is the fate of the zLL and its quantum Hall ground states. It was early predicted that if the Zeeman spin-splitting (enhanced by exchange interaction) overcomes the valley splitting, a topological inversion between the lowest electron-type and highest hole-type sub-levels should occur7,8. At charge neutrality, the ensuing ground state is a quantum Hall ferromagnet with two filled states of identical spin polarization, and an edge dispersion that exhibits two counter-propagating, spin-filtered helical edge channels (Fig. 1a,b), similar to those of the time-reversal symmetric 2D TIs6,36,37. Such a spin-polarized ferromagnetic (F) phase belongs to the recently identified new class of interaction-induced TIs with zero Chern number, termed quantum Hall topological insulators12 (QHTIs), which arise from a many-body interacting Landau level and can be pictured as two independent copies of quantum Hall

In graphene the immediate consequence of the Coulomb interaction is an instability towards quantum Hall ferromagnetism. Due to exchange interaction, a
FIG. 1. Spin-polarized ferromagnetic phase in graphene on high-$k$ dielectric. a, In the ferromagnetic phase of charge neutral graphene, the broken-symmetry states of the zeroth Landau level are spin-polarized and occupy both sublattices of the honeycomb lattice as shown in the top inset. The edge dispersion results from linear combinations of the bulk isospin states, which disperse as electron-like and hole-like branches, yielding a pair of counter-propagative, spin-filtered helical edge channels at charge neutrality.7,32 b, Schematics of a graphene lattice with helical edge channels propagating on the crystallographic armchair edge. c, Schematics of the hBN-encapsulated graphene device placed on a SrTiO$_3$ substrate that serves both as high-$k$ dielectric and back-gate dielectric. Due to the considerable dielectric constant of the SrTiO$_3$ substrate at low temperature and the ultra-thin hBN spacer (2−5 nm thick), Coulomb interaction in the graphene plane is significantly screened, resulting in a modification of the quantum Hall ground state at charge neutrality and the emergence of the ferromagnetic phase with helical edge transport. The inset shows the atomic layers of the hBN-encapsulated graphene van der Waals assembly, and the surface atomic structure of SrTiO$_3$.

systems with opposite chiralities, for which the sum of the individual Chern numbers is zero. Remarkably, unlike 2D TIs$^{6,36,37}$, immunity from back-scattering for the helical edge channels does not rely on the discrete time-reversal symmetry, conspicuously broken here by the magnetic field, but on the continuous U(1) axial rotation symmetry of the spin polarization.$^{12,27}$

The experimental situation is however at odds with this exciting scenario. A strong insulating state is systematically observed on increasing perpendicular magnetic field in charge-neutral, high-mobility graphene devices. The formation of the F phase is presumably hindered by some lattice-scale electron-electron and electron-phonon interaction terms, whose amplitudes and signs can be strongly renormalized by the long-range part of the Coulomb interaction$^{11}$, favoring various insulating spin- or charge-density-wave orders$^{9-11}$. Only with a very strong in-plane magnetic field component such that Zeeman energy overcomes the other anisotropic interaction terms does the F phase emerge experimentally.$^{26,27}$

Another strategy to engineer a F phase uses small-angle twisted graphene bilayers, with each layer sets in a different quantum Hall state of opposite charge polarity via a displacement field$^{38}$. Yet, those approaches realized hitherto suffer from either unpractical strong and tilted magnetic field or the complexity of the twisted layers assembly.

In this work we take a novel route to induce the F phase in monolayer graphene in a straightforward fashion. Instead of boosting the Zeeman effect with a strong in-plane field, we mitigate the effects of the lattice-scale interaction terms by a suitable substrate screening of the Coulomb interaction to restore the dominant role of the spin-polarizing terms and induce the F phase. We use a high-$k$ dielectric substrate, the quantum paraelectric SrTiO$_3$ known to exhibit a very large static dielectric constant of the order of $\epsilon \approx 10^4$ at low temperatures$^{39}$ (see Supplementary Figure S3), which acts both as an electrostatic screening environment and back-gate dielectric$^{40}$. For an efficient screening of the Coulomb potential, the graphene layer must be sufficiently close to the substrate, with a separation less than the magnetic length $l_B = \sqrt{\hbar/eB}$ ($\hbar$ the reduced Planck constant, $e$ the electron charge), which is the relevant length scale in the quantum Hall regime. High mobility hexagonal boron-nitride (hBN) encapsulated graphene heterostructures$^{41}$ were thus purposely made with an ultra-thin bottom hBN layer with a thickness, $d_{BN}$, ranging between
FIG. 2. Low magnetic field quantum spin Hall effect. a, Two-terminal resistance $R_{2t}$ in units of $\hbar/e^2$ of sample BNGrSTO-07 versus magnetic field $B$ and back-gate voltage $V_{bg}$ measured at 4 K. In addition to standard quantum Hall plateaus at filling fraction $\nu = 1$ and 2, the resistance exhibits an anomalous plateau around the charge neutrality point between $B = 1.5$ and 4 T, delimited by the black dotted lines and the arrow, which signals the regime of the QSH effect in this sample. The value of the resistance at this plateau is $\hbar/e^2$ and is color-coded in white. The inset schematics indicates the contact configuration. Black contacts are floating. The red and blue arrows on the helical edge channels indicates the direction of the current between contacts.b, Two-terminal conductance $G_{2t} = 1/R_{2t}$ in units of $e^2/h$ versus $V_{bg}$ extracted from a, at different magnetic fields. The first conductance plateaus of the quantum Hall effect at $2e^2/h$ and $6e^2/h$ are well defined. The QSH plateau of conductance $e^2/h$ clearly emerges at charge neutrality around $V_{bg} = 0$ V. c, Resistance at the charge neutrality point (CNP) versus $B$ for sample BNGrSTO-07 (red dots) extracted from a, and sample BNGrSTO-09 (blue dots). Whereas the sample with a thin hBN spacer exhibits a strong positive magnetoresistance at low $B$ diverging towards insolation, the sample with the thin hBN spacer shows the QSH plateau that persists up to $\sim 4$ T, followed by a resistance increase at higher $B$.

2–5 nm (see Fig. 1c, and Supplementary note 1) inferior to the magnetic length for moderate magnetic field (e.g. $l_B > 8$ nm for $B < 10$ T).

The emergence of the F phase in such a screened configuration is readily seen in Fig. 2a, which displays the two-terminal resistance of a hBN encapsulated graphene device in a six-terminal Hall bar geometry, as a function of the back-gate voltage $V_{bg}$ and magnetic field. Around the charge neutrality ($V_{bg} \approx 0$ V), an anomalous resistance plateau develops over a $B$-range from 1.5 to 4 T indicated by the two dotted black lines. This plateau reaches the quantum of resistance $\hbar/e^2$, color-coded in white. At $B > 5$ T, the resistance departs from $\hbar/e^2$ towards insolation as seen by the red color-coded magnetoresistance peak and shown in Fig. 2c.

The unusual nature of this resistance plateau can be captured with the line-cuts of the two-terminal conductance $G_{2t} = 1/R_{2t}$ versus $V_{bg}$ at fixed $B$, see Fig. 2b. While standard graphene quantum Hall plateaus at $G_{2t} = 4\pi^2/(N + \frac{1}{2}) = 2e^2/h$ and $6e^2/h$ for the Landau level indices $N = 0$ and $N = 1$ are well developed as a function of back-gate voltage, the new plateau at $G_{2t} = e^2/h$ is centered at the charge neutrality and does not show any dip at $V_{bg} = 0$ V. This behavior is at odds with the usual sequence of broken-symmetry states setting with magnetic field where first the insulating broken-symmetry state opens at $\nu = 0$ with $G_{2t} = 0$, followed at higher field by the plateaus of the broken-symmetry states at filling fraction $\nu = \pm 1$ (Ref.9,35). In Figure 2a, the latter states at $\nu = \pm 1$ arise for $B > 6$ T together with the insulating magnetoresistance peak at $\nu = 0$, that is, above the field range of the anomalous plateau. Hence, this striking observation of a $\hbar/e^2$ plateau at low magnetic field conspicuously points to a new broken-symmetry state at $\nu = 0$. We show in the following that this $\hbar/e^2$ plateau is a direct consequence of the spin-
polarized F phase and its helical edge channels.

Helical edge transport has unambiguous signatures in multi-terminal device configuration as each ohmic contact acts as a source of back-scattering for the counter-propagating helical edge channels with opposite spin-polarization. An edge section between two contacts is indeed an ideal helical quantum conductor of quantized resistance $\frac{h}{e^2}$. The two-terminal resistance of a device therefore ensues from the parallel resistance of both edges, each of them being the sum of contributions of each helical edge sections. As a result,

$$R_{2t} = \frac{h}{e^2} \left( \frac{1}{N_L} + \frac{1}{N_R} \right)^{-1}, \quad (1)$$

where $N_{L,R}$ is the number of helical conductor sections for the left (L) and right (R) edge between the source and drain contacts. By swapping the source and drain contacts for various configurations of $N_{L,R}$, one expects to observe resistance plateaus given by Eq. (1). Figure 3a displays a set of four different configurations of two-terminal resistances at $B = 2.5$ T as a function of back-gate voltage. Swapping the source and drain contacts and the number of helical edge sections (see contact configurations in Fig. 3b) yields a maximum around charge neutrality which reaches the expected values indicated by the dotted horizontal lines, thereby demonstrating helical edge transport. Notice that the plateau at $\frac{h}{e^2}$ in Fig. 2a is also fully consistent with Eq. (1) for $N_L = N_R = 1$.

Four-terminal non-local configuration provides another stark indicator for helical edge transport. Figure 3c shows simultaneous measurements of the two-terminal resistance between the two blue contacts (see sample schematics in the inset), and the non-local resistance $R_{NL}$ measured on the red contacts while keeping the same source and drain current-injection contacts. While $R_{2t}$ nearly reaches the expected value indicated by the dashed lines, namely $\frac{h}{e^2} \left( N_L = 5 \right.$ and $N_R = 1$), a non-local voltage develops in the $V_{bg}$ range that coincides with the helical edge transport regime in $R_{2t}$. The large value of this non-local signal, much larger than what could be expected in the diffusive regime, demonstrates that current is flowing on the edges of the sample. For helical edge transport, the expected non-local resistance is given by $R_{NLS} = R_{2t} \frac{N_L}{N_L}$, where $N_L$ and $N_V$ are the number of helical conductor sections between the source and drain contacts along the edge of the non-local voltage probes and between the non-local voltage probes, respectively. The measured $R_{NLS}$ shown in Fig. 3c is in excellent agreement with the expected value $\frac{h}{6 e^2} \left( N_L = 5 \right.$ and $N_V = 1$) indicated by the dashed line.

This global set of data that are reproduced on several samples (see Supplementary Note 3) therefore provides compelling evidence for helical edge transport, substantiating the F phase as the ground state at charge neutrality of substrate-screened graphene.

To assess the robustness of helical edge transport we conducted systematic investigations of its temperature, $T$ and magnetic-field dependences. Figure 4c displays a color-map of the two-terminal resistance of sample BNGrSTO-07 measured at charge neutrality as a function of magnetic field and temperature. The expected resistance value for the contact configuration shown in the inset schematics is $R_{2t} = \frac{2}{3} \frac{h}{e^2}$. This quantized resistance value is matched over a remarkably wide range of temperature and magnetic field, which is delimited by the dashed black line, confirming the metallic character of the helical edge transport. To ascertain the limit of quantized helical edge transport we measured different contact configurations at some bordering magnetic field and temperature values (see Supplementary Note 4), indicated in Fig. 4c by the green and red stars for quantized and not quantized resistance, respectively. The temperature and magnetic field dependences are further illustrated by Fig. 4a and b, which show the two-terminal resistance (measured in a different contact configuration, see inset Fig. 4b) versus back-gate voltage and the resistance at the charge neutrality point versus $B$, respectively, for various temperatures.

These data show that quantized helical edge transport withstands very high temperatures, up to 110 K, with an onset at $B \sim 1$ T virtually constant in temperature. Such a broad temperature range is comparable to WTe$_2$ for which QSH effect was observed in 100 nm-short channels up to 100 K (Ref.37). The new aspect of the F phase of graphene is that the helical edge channels formed by the broken-symmetry states retain their topological protection over large distances at elevated temperatures, namely 1.1 $\mu$m for the helical edge section between contacts of the sample measured in Fig. 4a-c. Various mechanisms can account for the high temperature breakdown of the helical edge transport quantization, such as activation of bulk charge carriers or inelastic scattering processes that break the U(1) spin-symmetry of the QHTI. As the former would reduce resistance by opening conducting bulk channels, the upward resistance deviation upon increasing $T$ rather points to inelastic processes that do not conserve spin-polarization. Consequently, this suggests that quantized helical edge transport may be retained at even higher temperatures for lengths below 1 $\mu$m.

Interestingly, the high magnetic field limit in Fig. 4c is temperature dependent. The lower the temperature the earlier in magnetic field start deviations from quantization: At $T = 4$ K, we observe an increase of resistance on increasing $B$ from about 3 T (see Fig. 2a and c and Fig. 4a-c), whereas this limit moves to 11 T at $T = 80$ K. For $B \gtrsim 3$ T, the resistance exhibits an activated insulating increase with lowering temperature, with a corresponding activation energy that increases linearly with $B$ (see Fig. 4e, red points. Data are taken on a different sample exhibiting an onset to insulation at $B \approx 7$ T). Such a behaviour indicates a gap opening in the edge excitation spectrum as illustrated in the inset Fig. 4d schematics, breaking down the helical edge transport at low temperature. This linear $B$-dependence of the activation energy
FIG. 3. Non-local helical edge transport. 

**a:** Two-terminal resistance versus back-gate voltage measured at 2.5 T for different contact configurations schematized in **b**. The inset shows an optical picture of the measured sample BNGrSTO-07. The scale bar is 4 μm. Each contact configuration yields a resistance at charge neutrality reaching the expected values for helical edge transport, which are indicated with the horizontal dashed lines. **b:** Schematics of the measurement configurations. Black contacts are floating. The red and blue arrows on the helical edge channels indicates the direction of the current between contacts. 

**c:** Two-terminal resistance, $R_{2t}$, in blue and non-local, four-terminal resistance, $R_{NL}$, in red versus back-gate voltage, in the contact configuration shown in the inset schematics. 

**d:** Resistance at the charge neutrality point (CNP), $V_{bg} = 0$, in the same contact configuration as in **c**, versus magnetic field. The helical plateau is observed for both two- and four-terminal resistances between 1 T and about 6 T.

...further correlates with the high magnetic field limit of the helical edge transport in Fig. 4c, thereby explaining why the limit for quantized helical edge transport increases to higher magnetic field with $T$.

The origin of the gap in the edge excitation spectrum is most likely rooted in the enhancement of correlations with magnetic field. An interaction-induced topological quantum phase transition from the QHTI to one of the possible insulating, topologically trivial quantum Hall ground states with spin or charge density wave order is a possible scenario\textsuperscript{12}. Such a transition is expected to occur without closing the bulk gap\textsuperscript{12,27}, which we confirmed via bulk transport measurements performed in a Corbino geometry (see Supplementary Note 5). Yet the continuous transition involves complex spin and isospin textures at the edges due to the $B$-enhanced isospin anisotropy\textsuperscript{43}, yielding the edge gap detected in Hall bar geometry. Another scenario relies on the helical Luttinger liquid\textsuperscript{44} behaviour of the edge channels, for which a delicate interplay between $B$-enhanced correlations, disorder and coupling to bulk charge-neutral excitations may also yield activated insulating transport\textsuperscript{45}.

To firmly demonstrate the key role of the SrTiO$_3$ dielectric substrate in the establishment of the F phase, we conducted identical measurements on a sample made with a 60 nm thick hBN spacer, much thicker than $l_B$ at the relevant magnetic fields of this study, so that screening by the substrate is irrelevant in the quantum Hall...
Resistance at CNP (kΩ)

Magnetic field (T)

Temperature (K)

FIG. 4. Phase diagram of the helical edge transport. a, Two-terminal resistance of sample BNGrSTO-07 versus back-gate voltage measured at different temperatures and a magnetic field of 4 T. The back-gate voltage is renormalized to compensate the temperature-dependence of the substrate dielectric constant (see Supplementary Note 8). b, Two-terminal resistance at the charge neutrality point (CNP) for the same data as in a. The inset shows the contact configuration used in a and b. c, Two-terminal resistance at the CNP versus magnetic field and temperature for a different contact configuration shown in the inset. The resistance shows a plateau at the value expected for helical edge transport, that is, $R_{\text{CNP}}(B,T) = 0$ (see blue dots in Fig. 4e, and Supplementary Note 6), as expected for a charge excitation gap that scales as $\Delta = 0.4\epsilon_C$, and the intercept $\Gamma = 27$ K describes the disorder-broadening of the Landau levels, which is consistent with the sample mobility (see Supplementary Note 1).

regime. Shown in Fig. 2c: with the blue dots, the resistance at the charge neutrality point diverges strongly upon applying a small magnetic field, thus clearly indicating an insulating ground state without edge transport. Systematic study of the activated insulating behaviour leads to an activation gap that grows as $\sqrt{B}$ (see blue dots in Fig. 4e, and Supplementary Note 6), as expected for a charge excitation gap that scales as the Coulomb energy $\epsilon_C = e^2/4\pi\epsilon_0\epsilon_{\text{BN}}l_B$ where $\epsilon_0$ and $\epsilon_{\text{BN}}$ are the vacuum permittivity and the relative permittivity of hBN. This self-consistently demonstrates that the F phase emerges as a ground state due to a significant reduction of the electron-electron interactions by the high-$\kappa$ dielectric environment.

Understanding the substrate-induced screening effect for our sample geometry requires electrostatic considerations that take into account the ultra-thin hBN spacer between the graphene and the substrate (see Supplementary Note 7). The resulting substrate-screened Coulomb energy scale $\tilde{\epsilon}_C = \epsilon_C \times S(B)$ is suppressed by a screening factor $S(B) = 1 - \frac{\epsilon_{\text{STO}} - \epsilon_{\text{BN}}}{\epsilon_{\text{STO}} + \epsilon_{\text{BN}}} \frac{L_B}{\sqrt{l_B + 4d_{\text{BN}}}}$ where $\epsilon_{\text{STO}}$ is the relative permittivity of SrTiO$_3$. As a result, electrons in the graphene plane are subject to an unusual $B$-dependent screening that depends on the ratio $l_B/d_{\text{BN}}$ and is most efficient at low magnetic field (see Supplementary Fig. S11). Importantly, despite the huge dielectric constant of SrTiO$_3$ of the order of $\epsilon_{\text{STO}} \approx 10^4$ (see Supplementary Fig. S3), $\tilde{\epsilon}_C$ is scaled down by a factor 10 for $l_B/d_{\text{BN}} = 4$ due to the hBN spacer, which is still a significant reduction of the long-range Coulomb interaction.

How such a screening affects the short-range, lattice-scale contributions of the Coulomb and electron-phonon interactions that eventually determine the energetically favorable ground state is a challenging question since
the hBN spacer precludes screening at the lattice scale. While theoretical estimates of these anisotropy terms initially point to the spin-polarized F ground state, renormalization effects due to the long-range part of the Coulomb interaction\textsuperscript{46,47} result in anisotropy terms that can change signs and amplitudes in an unpredictable fashion\textsuperscript{11}. The insulating behaviour commonly observed in usual samples still points to a canted antiferromagnetic order at charge neutrality, bearing out the strong renormalization of the anisotropy terms. We conjecture that the reduction of the Coulomb energy scale by the substrate screening is the key that suppresses the renormalization effects, restoring the F phase as the ground state at charge neutrality. Therefore, enhancing the renormalization effects, restoring the F phase as the ground state at charge neutrality.

Finally, our work demonstrates that the F phase in screened graphene, which emerges at low magnetic field, provides a prototypical, interaction-induced topological phase, exhibiting remarkably robust helical edge transport in a wide parameter range. Interestingly, the role of correlations in the edge excitations, which are tunable via the magnetic field and an unusual B-dependent screening, should be of fundamental interest for studies of zero-energy modes in superconductivity-proximitized architectures constructed on the basis of helical edge states. We further expect that substrate-screening engineering, tunable via the hBN spacer thickness, could have implications for many other correlated 2D systems for which the dielectric environment drastically impact their ground states and (opto)electronic properties.

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METHODS

Sample fabrication

hBN/graphene/hBN heterostructures were made from exfoliated flakes using the van der Waals pick-up technique\textsuperscript{41}. Contacts were patterned by electron-beam lithography and metalized by e-gun evaporation of a Cr/Au bilayer after etching the stack directly through the resist pattern used to define the contacts. For sample BNGrSTOVH-02, a Hall-bar was first patterned by etching the stack using a CHF\textsubscript{3}/O\textsubscript{2} plasma and a mask of hydrogen silsesquioxane (HSQ) resist. Contacts were then designed and deposited on the etched Hall-bar with apparent graphene edges. The design and contacting process of the sample in a Corbino geometry (BNGrSTOCorbino1) is detailed in the Supplementary Note 5. For all samples we used 500\(\mu\)m thick SrTiO\(_3\) (100) substrates that are cleaned with hydrofluoric acid buffer solution before deposition of the hBN/graphene/hBN heterostructures.

Measurements

Two and four-terminals measurements were performed with a standard low-frequency lock-in amplifier technique in voltage-bias configuration by applying an ac bias voltage of 0.4 mV. To compensate for the dielectric hysteresis of the SrTiO\(_3\) substrate, back-gate dependence of the resistance was systematically carried out with the same back-gate voltage sweep. This enabled to obtain reproducible position of the charge neutrality point in back-gate voltage. For the sake of clarity, the back-gate voltage axes of all figures are shifted to have the charge neutrality point at zero voltage.

(2009).


