

Evidence for a spontaneous gapped state in ultraclean bilayer graphene

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At the charge neutrality point, bilayer graphene (BLG) is strongly susceptible to electronic interactions and is expected to undergo a phase transition to a state with spontaneously broken symmetries. By systematically investigating a large number of single- and double-gated BLG devices, we observe a bimodal distribution of minimum conductivities at the charge neutrality point. Although σ_{\min} is often approximately $2\text{--}3 e^2/h$ (where e is the electron charge and h is Planck's constant), it is several orders of magnitude smaller in BLG devices that have both high mobility and low extrinsic doping. The insulating state in the latter samples appears below a transition temperature T_c of approximately 5 K and has a $T = 0$ energy gap of approximately 3 meV. Transitions between these different states can be tuned by adjusting disorder or carrier density.

topological states | anomalous hall | spontaneous quantum Hall states | electron–electron interactions | layer antiferromagnets

Bilayer graphene (BLG) has provided a fascinating new platform for both post-silicon electronics and exotic many-body physics (1–23). Because its conduction and valence bands touch at two points in momentum space and have approximately quadratic dispersion accompanied by momentum-space pseudospin textures with vorticity $J = 2$, charge-neutral BLG is likely to have a broken-symmetry ground state in the absence of disorder (6–11, 15–18, 24–26). Theoretical work on the character of the ground state in neutral BLG has examined a variety of distinct but related pseudospin ferromagnet states—including gapped anomalous Hall (5, 6, 19), layer antiferromagnetic (6–8, 16–18, 22), and current loop states (26)—that break time-reversal symmetry, and gapless nematic states, which alter Dirac point structure and reduce rotational symmetry (6–11, 15–18, 24, 25). The pseudospin degree of freedom reflects the presence of two low-energy carbon sites per unit cell that are localized in different layers. Experimental work has confirmed the strong role of interactions, but has been equivocal in specifying ground-state properties. In particular, both gapped and gapless states have been reported (19–23) in suspended BLG. The low-temperature minimum conductivity at the charge neutrality point (CNP), σ_{\min} , has ranged from approximately 0.05 to 250 μS . These orders-of-magnitude differences between σ_{\min} values measured in apparently similar samples have been baffling.

In this paper we attempt to shed light on these ambiguous findings by systematically examining a large number of single- and double-gated BLG samples, with mobility values ranging from 500 to 2,000 $\text{cm}^2/\text{V}\cdot\text{s}$ for substrate-supported samples and 6,000 to 350,000 for suspended samples. We find a surprisingly constant σ_{\min} value of approximately $2\text{--}3 e^2/h$ for a majority of the devices (here, e is the electron charge and h is Planck's constant), independent of their mobility and of the presence or absence of substrates. However, for T below approximately 5 K, the best devices form an insulating state with an energy gap of approximately $2\text{--}3$ meV. Importantly, the transition between conducting and insulating states can also be tuned by varying charge

density n and perpendicular electric field E_{\perp} , in agreement with theoretical predictions for gapped pseudospin ferromagnetic states. Finally, our observation of a bimodal distribution of σ_{\min} values suggests that transport in the conducting devices could occur along domain boundaries that separate regions with different pseudospin order configurations.

Results

We fabricate single-gated BLG devices using a lithography-free technique and suspend double-gated BLG by combining acid etching with a multilevel lithographic technique to make devices with suspended top gates (*SI Text*). All suspended BLG devices as fabricated have relatively low mobilities, presumably due to gas or water absorption on the surface of BLG exposed to an ambient environment. Current annealing is performed in vacuum (Fig. 1C). The optimal state is normally achieved when I starts to saturate (*SI Text*) at approximately 0.2 mA/ μm per layer.

Fig. 1D and E plot the two-terminal differential conductivity $\sigma = (L/W)dI/dV$ of two suspended BLG devices vs. back-gate voltage V_{bg} at $T = 1.5$ K after current annealing. (Here, L/W is the aspect ratio of the devices.) Both curves are steeply V shaped and have CNPs (marked by conductivity minima) that are close to $V_{\text{bg}} = 0$ V. Surprisingly, the σ_{\min} values of the two devices are drastically different: 2.5 and 0.02 e^2/h , respectively. The insulating behavior of the latter device is confirmed by current–voltage I – V_{sd} curves. In a magnetic field B , both devices display quantum Hall plateaus with the eightfold degeneracy (12, 13) of the zero-energy Landau level (LL) fully lifted (*SI Text*). From the Landau fan diagram that plots the differential conductance G (color) vs. V_{bg} and B (Fig. 1D and E, *Insets*), the $\nu = 0$ state is visible for both devices at $B > 0.5$ T and persists down to $B = 0$ for the device with very low σ_{\min} (21, 22).

To shed light on the origin of the large range of σ_{\min} values, we investigated nine substrate-supported BLG devices and 23 suspended BLG devices with aspect ratios between 0.5 and 2, and areas from 1 to 18 μm^2 . The results are summarized in Fig. 2A, which plots σ_{\min} as a function of field-effect mobility $\mu = \frac{1}{e} \frac{d\sigma}{dn}$ for each device. Evidently, the data points separate into two groups. Most data points fall into group I, in which σ_{\min} is almost independent of mobility and similar for suspended and supported devices. Within this class of devices, the CNP conductivity is approximately 100 μS or approximately $2.8 e^2/h$ (27–33).

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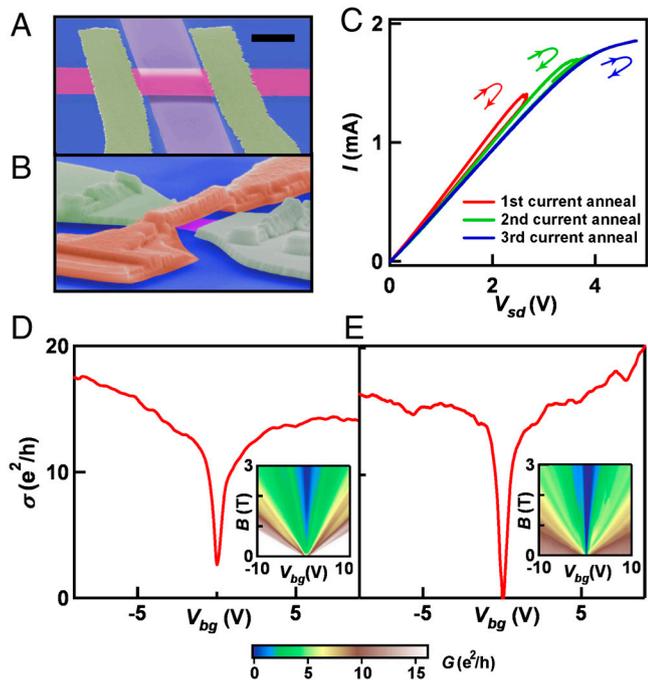


Fig. 1. (A, B) False color-scanning electron micrograph of BLG device with and without top gate (Scale bar: 2 μm). (C) I - V_{sd} curves of a suspended BLG during current annealing. (D, E: Main and Insets) $\sigma(V_g)$ and $G(V_{bg}, B)$ for two BLG devices with and without insulating state at CNP ($T = 1.5$ K).

Very different behavior is found in the seven devices that fall into group II: σ_{\min} is at most $0.4 e^2/h$, and as low as $1 \mu\text{S}$. Notably, all seven devices have very high mobility. To identify the physical difference between the two classes of devices, we also examined V_{CNP} (the V_{bg} required to reach the CNP, which is a proxy for the overall doping level). Fig. 2 B and C plots σ_{\min} and μ vs. V_{CNP} for all suspended samples, with the insulating devices denoted by blue triangles. Two striking features are evident: (i) μ decreases

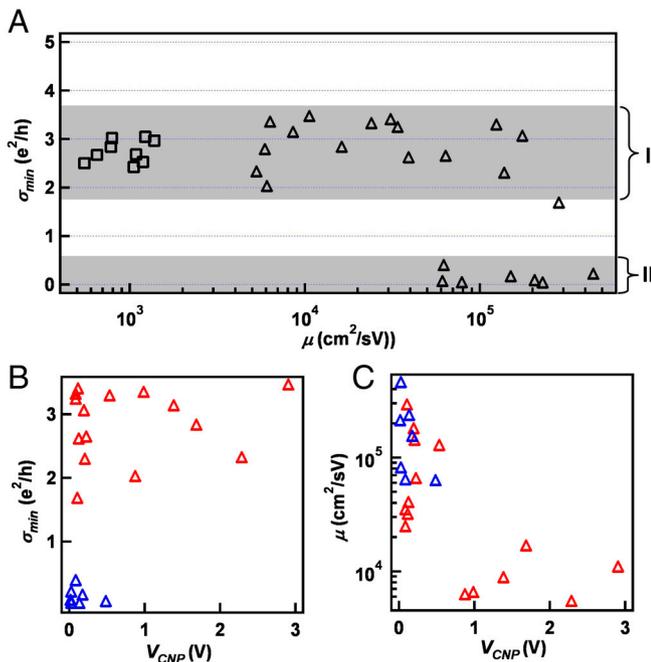


Fig. 2. (A) $\sigma_{\min}(\mu)$ for ninesubstrate-supported BLG devices (square symbols) and 23 suspended BLG devices (triangular symbols) at 1.5 K (except for three devices in region II, which was taken at $T = 0.3$ K). (B, C) $\mu(V_{\text{CNP}})$ and $\sigma_{\min}(V_{\text{CNP}})$ for suspended BLG devices. The blue symbols denote devices in region II.

with increasing V_{CNP} in agreement with previous reports in substrate-supported graphene (34, 35), suggesting that charged impurities are important scatterers even in these high-mobility devices; and (ii) the insulating BLG devices in Fig. 2 B and C cluster around $V_{\text{CNP}} = 0$. Insulating behavior at the CNP is observed only in devices with both high mobility and low charged-impurity density.

To obtain further insight we compare the temperature dependences of group I and group II devices. Fig. 3A displays σ_{\min} on a logarithmic scale vs. $1/T$ for $1.4 \leq T \leq 100$ K for one non-insulating device and two different insulating BLG devices. The inset plots the same data sets $\sigma_{\min}(T)$ on linear-log scales. Amazingly, for $10 < T < 100$ K, the $\sigma_{\min}(T)$ curves of all three devices collapse into a single curve. This is in contrast with previous work on single-layer (36) and trilayer graphene (37, 38), which reported large sample-to-sample variation in $\sigma_{\min}(T)$. The consistent behaviors among three devices for $T > 10$ K strongly suggest that we are indeed observing intrinsic attributes of BLG.

The behaviors of the device classes start to deviate at approximately 5–7 K: σ_{\min} of the non-insulating device decreases only modestly; in contrast, the σ_{\min} of both insulating devices exhibits an abrupt change in slope and drops precipitously for $T < 5$ K where the data are well described by $\sigma_{\min}(T) = A \exp(-E_A/2 k_B T)$. (Here, A is the prefactor, E_A is the activation energy, and k_B is the Boltzmann's constant). The best fit is obtained by using $A = 17 e^2/h$ and E_A/k_B of approximately 18 K, indicating thermally activated transport over a gap $E_A = 1.6$ meV.

These data suggest the presence of a gap in insulating devices for $T < 5$ K. To investigate this further, we study σ vs. source-drain bias V_{sd} at the CNP (21, 22) (Fig. 3B). At $T = 1.4$ K, σ increases precipitously when $|V_{sd}|$ increases from 0, forming a U-shaped profile and reaching two dramatic peaks at ± 2.8 mV, and decreases again to approximately $8 e^2/h$ for $|V_{sd}| > 5$ mV. These $\sigma(V_{sd})$ curves resemble the density of states of gapped phases, like superconductors, charge density waves, and, perhaps most pertinently, the displacement field-induced gapped BLG state (4, 39–41). Because the device has symmetric coupling to both electrodes, we take the magnitude of the gap to be half of the separation between the two peaks (approximately 2.8 meV). This is larger than the value of approximately 1.6 meV obtained from thermal-activation measurements, possibly reflecting the contribution of variable range hopping and other disorder-related effects to the temperature dependence of transport. Thus, the $\sigma(V_{sd})$ curves, together with the $\sigma_{\min}(T)$ measurements, unequivocally establishes the presence of a low-temperature gap of approximately 2–3 meV in the charged excitation spectrum of insulating devices.

We now examine the $\sigma(V_{sd})$ curves of the insulating devices at different temperatures (Fig. 3B). When T increases from 1.4 K, σ_{\min} increases, $\sigma(V_{sd})$ adopts a V-shaped profile, and the magnitudes of the two peaks decrease and vanish entirely at approximately 5 K. These observations suggest the disappearance of the gap for $T > 5$ K. Our data thus provide strong evidence for a finite temperature phase transition to a gapped state with a critical temperature T_c of approximately 5 K and a gap Δ/k_B (approximately 20–30 K). The temperature dependence of the gap strongly suggests that it is of many-body origin, and the rough correspondence between the critical temperature and gap scales suggests that the broken symmetry is reasonably well described by mean-field theory (SI Text).

Our data thus far suggest a T -dependent phase transition in charge-neutral BLG between a conducting state and an interaction-induced insulating state. The conducting state could be due to bulk two-dimensional metallic behavior, or, alternatively, due to transport along topologically protected edge states supported by domain walls separating regions (5, 6) with different spin-and valley-dependent Chern numbers. Future experiments will be

from a metallic state to an insulating state with an energy gap of approximately 2–3 meV. The latter arise from electronic correlation and are likely spontaneous quantum Hall states *without* overall layer polarization (6, 22). Interestingly, increasing n or disorder has an effect that is similar to temperature on the insulating state, suggesting that these parameters can tune continuous quantum phase transitions. Increasing E_{\perp} with either polarity will also induce a transition into a topologically different gapped state *with* layer polarization. We expect that the rich physics we have studied in BLG and the closely related phenomena that are expected in rhombohedral (ABC) stacked multilayer graphene (6, 18, 38, 41, 44) will provide considerable scope for future work.

Materials and Methods

Exfoliated BLG sheets are obtained by mechanical exfoliation and identified by color contrast in optical microscopy and Raman spectroscopy. Two different types of BLG devices are fabricated: (i) Graphene sheets are exfoliated on substrates or across predefined trenches that are 250 nm deep and approximately 3 μm wide, then coupled to 5-nm/50-nm Ti/Al metal electrodes through shadow mask evaporation (42). The typical back-gate coupling ratio of such devices is approximately $2.5 \times 10^{10} \text{ cm}^{-2} \text{ V}^{-1}$. Because of the lithography-free fabrication process, these devices are extremely clean, with

mobilities up to 450,000 $\text{cm}^2/(\text{Vs})$. (ii) BLG devices are fabricated with suspended top gates (43) using electron-beam lithography, and completed devices are released from the substrate by hydrofluoric-acid etching. These double-gated devices allow independent adjustment of induced charge density n and perpendicular electric field E_{\perp} . After fabrication we transfer our suspended BLG devices into a high-vacuum cryostat, and current annealing is performed at 1.5 K. Details of current-annealing procedure and the current-voltage characteristics at different stages of annealing are described in detail in *SI Text*.

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