Transport spectroscopy of symmetry-broken insulating states in bilayer graphene

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Bilayer graphene is an attractive platform for studying new two-dimensional electron physics1–5, because its flat energy bands are sensitive to out-of-plane electric fields and these bands magnify electron-electron interaction effects. Theory6–16 predicts a variety of interesting broken symmetry states when the electron density is at the carrier neutrality point, and some of these states are characterized by spontaneous mass gaps, which lead to insulating behavior. These proposed gaps6,7,10 are analogous7,18 to the masses generated by broken symmetries in particle physics, and they give rise to large Berry phase effects8,9 accompanied by spontaneous quantum Hall effects9,20. Although recent experiments21–25 have provided evidence for strong electronic correlations near the charge neutrality point, the presence of gaps remains controversial. Here, we report transport measurements in ultraclean double-gated bilayer graphene and use source-drain bias as a spectroscopic tool to resolve a gap of $\sim 2$ meV at the charge neutrality point. The gap can be closed by a perpendicular electric field of strength $\sim 15$ mV nm$^{-1}$, but it increases monotonically with magnetic field, with an apparent particle-hole asymmetry above the gap. These data represent the first spectroscopic mapping of the ground states in bilayer graphene in the presence of both electric and magnetic fields.

The single-particle band structure of bilayer graphene resembles that of a gapless semiconductor, with parabolic valence and conduction bands touching at the highly symmetric $K$ and $K'$ Dirac points. When weak remote hopping processes are included, the momentum space band-touching point splits into four4, and Lifshitz transitions occur at low carrier densities (for discussion see Supplementary Information). For a perpendicular electric field $E_{\perp} \neq 0$, a bandgap develops and increases with $E_{\perp}$, saturating at $\sim 0.3$ eV (refs 4,26,27), and the conduction band has a ‘Mexican hat’ shape. When electron–electron interactions are included, bilayer graphene is expected to be unstable to broken-symmetry states, which can be viewed as layer-pseudospin ferromagnets6.

Bilayer graphene has been described using a two-band model8 that is valid near the charge neutrality point with the broken-symmetry state quasi-particle Hamiltonian

$$H = -\left(\frac{p^2}{2m^*}\right)\left[\cos(2\phi_p)\sigma_0 \pm \sin(2\phi_p)\sigma_z\right] - \Delta \cdot \sigma$$  \hspace{1cm} (1)

The first term in equation (1) is the quadratic band Hamiltonian, where $\tan\phi_p = p_y/p_x$, where $p$ is the angular momentum, $m^*$ is the effective mass of the carriers, $\sigma$ is a Pauli matrix vector that acts on the layer degree of freedom, $\Delta$ represents the order parameter of the broken symmetry state, and the $\pm$ signs refer to the $K$ and $K'$ valleys, respectively. It has been variously predicted6–10 to be oriented in the $\pm z$-direction, yielding gapped isotropic states with large momentum-space Berry curvature8, or in the $x$–$y$ plane, yielding gapless anisotropic (nematic) states11,12 with vanishing Berry curvatures. A variety of distinct but related massive states occur, depending on the relation of the sign of $\Delta_0$ to spin and valley6. The attributes of some theoretically proposed bilayer graphene states are summarized in Table 1 and Fig. 1, and the relationship between $\Delta$ and quasiparticle electronic properties is discussed in detail in the Supplementary Information.

In the first experimental reports21–25, which hinted at an ordered state in bilayer graphene, the minimum conductivity $\sigma_{\text{min}}$ was a non-monotonic function of $E_{\perp}$ with a value $\sim 60$ $\mu$S at $n = E_{\perp} = 0$, although the experimental evidence for spontaneous gaps in this work was not conclusive. Recent works using single-gated samples reported either very high $\sigma_{\text{min}} \approx 7e^2/h$ (where $e$ is electron charge and $h$ is Planck’s constant), which was attributed to the gapless nematic phase25, or low $\sigma_{\text{min}} \approx 0.3e^2/h$, which hinted at a gapped state23. Thus, the nature of the ground state at the charge neutrality point remains very controversial. Here, we demonstrate that the ground state is indeed insulating and gapful with a magnetic field $(B)$-dependent gap of the form

$$E_{\text{gap}} = \Delta_0 + \sqrt{a^2B^2 + \Delta_0^2}$$  \hspace{1cm} (2)

where $|\Delta_0| \approx 1$ meV and $a \approx 5.5$ meV T$^{-1}$. By tracking the dependence of $E_{\text{gap}}$ on $B$ and on $E_{\perp}$, which is believed to induce a transition between a layer-unpolarized and a layer-polarized state, we are able to provide a spectroscopic mapping of bilayer graphene ground states.

Our devices consist of exfoliated bilayer graphene sheets with chromium/gold electrodes suspended between silicon/SiO$_2$ back gates and metal top gates (Fig. 1a). The devices’ field effect mobility $\mu_{FE} = 1/(e\langle d\sigma/dn\rangle)$, where $\sigma$ is the device conductivity, typically calculated by taking the slope of the $\sigma(V_{bg})$ curves between $n = 0$ and $n \approx 4 \times 10^{10}$ cm$^{-2}$, is $\sim 80,000–100,000$ cm$^2$ V$^{-1}$ s$^{-1}$. By tuning the voltages applied to the back-gate $V_{bg}$ and top-gate $V_{tp}$, we can independently control $E_{\perp}$ and the charge density $n$ induced in the bilayer. We find that this capability is of utter importance, as even a small amount of $E_{\perp}$ or $n$, which may be inadvertently present in single-gated devices, can obscure the insulating state and significantly elevate $\sigma_{\text{min}}$ (see Supplementary Information).

Figure 1c plots the two-terminal differential conductance $G = dI/dV$ at $V = 0$ of the device (colour) versus $V_{bg}$ and $B$ and reveals Landau levels as coloured bands radiating from the charge neutrality point and $B = 0$. Line traces of $G(V_{bg})$ at constant $B$ exhibit conductance plateaus with values near 0, 1, 2, 3, 4 and
8e/h (Fig. 1d), indicating that the eightfold degeneracy of the lowest Landau level (LL) is broken. Observation of these plateaux at relatively low B underscores the high quality of the device.

Close inspection of Fig. 1d reveals that the v = 0 gap appears to persist down to B = 0. Indeed, at B = 0, the G(E⊥n) plot shows a local minimum at n = E⊥ = 0 (Fig. 2a,b). This contradicts the single-particle picture, which predicts a gap that is roughly linear in E⊥, hence a monotonically decreasing G(E⊥). Our data therefore suggest a breakdown of the non-interacting electron picture, even allowing for the possibility of uncontrollable mechanical deformations in our suspended flakes (Supplementary Information).

To investigate the resistive state at the charge neutrality point spectroscopically, we measure G as a function of source–drain bias V and E⊥ while keeping n = 0. Typical spectroscopic transport measurements are performed using tunnel probes, but our devices have highly transparent contacts. Because they are in the quasi-ballistic limit, we nevertheless anticipate spectroscopic resolution. Our nonlinear transport data are summarized in Fig. 2c–f. The most striking feature is the region of dark blue/purple at the centre of the plots, corresponding to the highly resistive state at small E⊥. At n = E⊥ = V = 0, the device is insulating, with Gmin ≈ 0.5 μS (Fig. 2f, green curve). We note that this insulating state is observed only in high-mobility samples realized after current annealing, and then only at low sweeping rates (Supplementary Information) and after careful optimization of the measurement set-up to eliminate spurious voltage noise. As V increases, G remains approximately 0 until it increases abruptly at V ≈ ±1.9 mV, reaching sharp peaks before it decreases again to ~300 μS. The G(V) curve bears a striking resemblance to the tunnelling density of states of a gapped insulator, and strongly suggests the formation of an ordered phase with an energy gap Egap ≈ 1.9 meV. This value is further corroborated by temperature-dependent measurements of σmin for a sample in the gapped regime, and by the n-dependence of the gap (Supplementary Information). Importantly, this gap can be closed by application of E∥ of either polarity; G increases with |E∥| (Fig. 2d), and upon application of moderate E∥ > ~15 mV nm−1, the gap-like structure completely vanishes and the G(V) curve becomes approximately V-shaped, with a finite conductance minimum of ~100 μS at n = 0 (Fig. 2f, purple curve). Finally, for sufficiently large E∥, G(V = 0) begins to decrease with increasing E∥ (Fig. 2d), reverting to single-particle behaviour.

Such an insulating state, which is the most salient experimental feature, is only observed in devices with the highest mobility. To gain further insight, we studied its evolution with B (Fig. 3). When B increases from 0, the gapped phase continuously evolves into a bilayer insulating state at filling factor v = 0, with the sharp peaks in G becoming sharper and more dramatic. The magnitude of the gap, as measured from the bias values of the sharp peaks in G, is well-described by equation (2), where |Δn| ≈ 1 meV (obtained from Fig. 2f) and a = 5.5 mV T−1. For B > 0.5 T, Egap ≈ 5.5B (meV T−1), which is much larger than the single-particle gap induced by Zeeman splitting, ~0.1 meV T−1. We note that our observation of a linear dependence of Δn on B is consistent with previous reports28,33, but with a significantly larger magnitude, possibly owing to the superior quality of our device. Another noteworthy feature of Fig. 3a is that the conductance peaks at positive and negative bias voltages, which we associate with conduction and valence band edges, are highly asymmetric. Because the current–voltage characteristics of the device are symmetric in V outside the gapped region, this observed asymmetry, which increases with increasing B and reverses when B changes sign, does not arise from contact asymmetry, nor can it be accounted for by the Onsager relation because the measurements are carried out in the nonlinear regime. Rather, it suggests particle–hole asymmetry in the device.

Our experimental findings may be summarized as follows: (i) ultraclean bilayer graphene is insulating at n = B = E⊥ = 0, with an energy gap Egap ≈ 1.9 mV that can be closed by E∥ of either sign; (ii) the energy gap evolves in B following equation (2); and (iii) this state is apparently particle–hole asymmetric. These observations provide much insight into the nature of bilayer graphene’s symmetry-broken ground state. For instance, observation (i) rules out gapless ordered states11,12. For the gapped states, the symmetric dependence on E∥ indicates that there is no net charge imbalance between the two layers, thus excluding states with net spontaneous layer polarization like the charge layer polarized (CLP) state depicted in Fig. 1. We note that the CLP phase, also called the quantum valley Hall (QVH) state, is expected to be the ground state under sufficiently large E⊥, as observed experimentally.

Thus, among the proposed states, we are left with the three gapped state candidates that have no overall layer polarization. Given the

| Table 1 | Attributes of possible ordered states in bilayer graphene at n = E⊥ = 0. |
|---------|-----------------|----------------|-----------------|-----------------|
| Gapped? | Nematic order    | QAH            | QSH             | LAF             | CLP (QVH) |
| No      | Yes             | 4e2/h          | Yes             | 0               | Yes       |
| Two-terminal σmin | Finite         | 4e2/h          | Yes             | 0               | Yes       |
| Broken symmetries | In-plane rotation | Time reversal; Ising valley | Spin rotational; Ising valley | Time reversal; spin rotation | Inversion |

QAH, quantum anomalous Hall; QSH, quantum spin Hall; LAF, layer antiferromagnet; CLP, charge layer polarized; QVH, quantum valley Hall.
Both QAH and QSH states are ruled out by their topologically single valley. If we assume ideal coupling of both layers to the considerably more difficult. Equation (2) applies to charges in a metricy-broken gapped phase in charge neutral graphene. The essential ingredient to form a layer antiferromagnet is electron rest mass, in good agreement with equation (2).

On the other hand, we note that because most of the metals are deposited on the top layer and contact the bottom layer only via the edges, the electrodes could couple preferentially to one layer (or equivalently, one valley). In this case, the LAF state, which is the only proposed insulating phase, can account for both observations (i) and (ii). Because the absence of edge states is the most robust experimental signature, our observations are most consistent with the LAF state, although the particle–hole asymmetry in $B$ is not explained within this picture. It is thus possible that a new ground state that has not been theoretically proposed underlies our observations. Further theoretical and experimental work will be necessary to ascertain the nature of the gapped state and achieve a full understanding of our observations.

Finally, we focus on the quantum Hall states in external electric and magnetic fields and further establish the bias-dependent measurement as a spectroscopical tool. Figure 4a plots $G$ versus $E_{\perp}$ and $n$ at constant $B = 3.5$ T. As shown by the line traces in Fig. 4b,c, at $E_{\perp} = 0$, only the $n = 0$ and $n = 4$ quantum Hall plateaux are observed; at finite $E_{\perp}$, all integer quantum Hall states between 0 and 4 are resolved, demonstrating degeneracy-lifting by the perpendicular electric field. From the spectroscopic data $G(V, E_{\perp})$ at constant $B$, the gap $\Delta_{\text{edge}}$ is diamond-shaped (Fig. 4d,e); its magnitude decreases linearly with applied $E_{\perp}$ of either polarity, until it is completely closed at a critical field $E_{\perp}^{*}$. Figure 4f plots $E_{\perp}^{*}$ obtained at three different $B$ values. The data points fall on a straight line, with a best-fit slope of $\sim 12.7$ mV nm$^{-1}$ T$^{-1}$. Extrapolation of the best-fit line to $B = 0$ yields a finite $E_{\perp}^{*}$ intercept $\sim 12.5$ mV nm$^{-1}$, which agrees with the critical $E_{\perp}^{*}$ value estimated from the zero $B$ field data in Fig. 2d. Both the slope and the finite $E_{\perp}^{*}$ intercept are consistent with those measured from the movement of the $n = 4$ plateau as functions of $E_{\perp}$ and $B$ (ref. 22; see Supplementary Information). Taken together, our data confirm that the bias-dependent measurement provides a spectroscopic
determination of the $v = 0$ gap, and indicates a transition between two different quantum Hall ferromagnetic phases. Interestingly, a recent theoretical analysis suggests the nature of these two phases is a canted antiferromagnetic and a CLP state. Such a finding is consistent with our previous conclusion of the LAF phase at $B = E_\text{F} = n = 0$.

In summary, we demonstrate the formation of a gapped, insulating phase in charge-neutral bilayer graphene. The gap is closed by application of a perpendicular $E$ field of either polarity and evolves into a $v = 0$ state in a magnetic field with a gap of $\sim 5.5$ meV $\text{T}^{-1}$, with apparent particle–hole symmetry. Our work, together with recent experiments, contributes towards understanding the rich interaction-driven physics in bilayer graphene. Further theoretical and experimental work, such as scanning tunnelling microscopy or optical Kerr effect measurements, is warranted to ascertain the nature of the gapped states.

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References


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Author contributions
C.N.L and J.V. conceived the experiments. Y.J. and P.K. isolated and identified the graphene sheets. R.S. assisted with sample preparation. J.V., L.J., W.B., Y.J. and D.S. performed transport measurements. C.N.L, M.B., W.B. and J.V. interpreted and analysed the data. V.A., C.V., F.Z., J.J. and A.H.M. interpreted data and performed theoretical calculations. C.N.L., J.V., F.Z., J.J. and A.H.M. co-wrote the paper. All authors discussed the results and commented on the manuscript.

Additional information
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