Canted antiferromagnetic to ferromagnetic phase transition in bilayer graphene

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Abstract. We performed electrical transport measurements in strong tilted magnetic fields (up to 30 T) on a high quality bilayer graphene sample. The possibility of tuning the system into a topological state at zero filling factor, associated to a ferromagnetic phase, is demonstrated on the basis of an insulator-to-metal transition and simultaneous arising of strong non-local response. Based on recent theoretical predictions, relevant aspects of the phase transition are quantitatively extracted. New experimental developments on the topics are proposed in the final section.

1. Introduction
The quantum spin Hall (QSH) effect is a new phase of matter where a gapped bulk coexist with gapless helical edge states, which are protected from backscattering by a symmetry of the physical system \cite{1, 2}. While time-reversal symmetry plays this role in two-dimensional topological insulators (2D TIs) such as inverted HgTe \cite{3, 4} and InAs/GaSb quantum wells \cite{5, 6}, an analogous topological phase is expected in single and double layer graphene at zero filling factor, provided that an intense in-plane component of the magnetic field (\(B_\parallel\)) strongly enhances the Zeeman splitting with respect to competing interaction anisotropies \cite{7, 8}. In particular, bilayer graphene is expected to be continuously tunable from a canted antiferromagnetic (CAF) to a ferromagnetic (F) state at the charge neutrality point (CNP), with associated closing of the energy gap for the one-dimensional edge channels and realization of two copies of the QSH effect \cite{9, 10, 11}.

2. Sample and experimental set-up
We studied this phenomenology in a van der Waals heterostructure \cite{12} made of a stacking of hexagonal boron nitride (h-BN)\textsuperscript{13}, bilayer graphene and h-BN flakes obtained by micro-mechanical cleavage. The sample, produced with the dry pick-up technique described in Ref.\textsuperscript{14}, was previously characterized with experiments both at zero magnetic field \textsuperscript{15} and at finite filling factor \textsuperscript{16}. We performed electrical transport measurements with standard low-frequency (\(\approx 13\))

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Hz) ac lock-in technique, using an excitation current of 100 nA and varying the carrier density with a dc back-gate voltage ($V_g$). The angle between the magnetic field and the graphene plane was varied continuously from 0° to 180°; the correct orientation of the sample was tested by carefully checking the carrier density corresponding to the integer quantum Hall states at filling factor ±4, which depends on the perpendicular component of the magnetic field ($B_\perp$) only.

3. Four-probe and non-local measurements in tilted magnetic fields

Figure 1 (a) shows the evolution of the four-probe resistance $R_{xx}$ as a function of $V_g$ when an increasingly strong $B_\parallel$ is applied in addition to a constant quantizing $B_\perp$. This is achieved by progressively increasing the tilting angle of the sample’s plane with respect to the field axis and consequently adjusting the total intensity of the field ($B_{\text{tot}}$). As $B_\parallel$ grows, the resistance at the CNP ($R_{CNP}^{xx}$) drops dramatically, eventually evolving into a local minimum. The suppression of $R_{CNP}^{xx}$ is indicative of the creation of new conduction channels at charge-neutrality. When the highest values of $B_\parallel$ are considered, the local minimum at the CNP assumes values $\frac{1}{4} \hbar/e^2 < R_{CNP}^{xx} < \frac{1}{3} \hbar/e^2$. The lower limit $\frac{1}{4} \hbar/e^2$ represents the theoretical value for the four-probe resistance in the F phase of bilayer graphene, which is calculated taking into account that six ohmic contacts to the graphene were employed in the present experiment and that each edge path between electrical contacts corresponds to a resistor of value $\frac{1}{2} \hbar/e^2$. The effect of the strong $B_\parallel$ applied is therefore consistent with the formation of four helical edge states, i.e. with the realization of two copies of the QSH effect. The not exact quantization detected in our measurements (also observed in Ref.[10] for bilayer graphene, Ref.[8] for single-layer graphene and Ref.[4, 3] for 2D TIs) indicates residual backscattering, likely to be due to short-circuiting via bulk states and/or non-ideal contacts acting as uncontrolled de-coherence sites [4].

Figure 1. (a) $R_{xx}$ as a function of $V_g$ for increasing $B_\parallel$ with fixed $B_\perp = 5$ T, at $T \simeq 1.2$ K. (b) Non-local resistance as a function of $V_g$ at $B_\perp = 7$ T, $T \simeq 1.2$ K, for increasing values of $B_\parallel$. The corresponding contacts’ configurations are shown in (c) for $R_{xx}$ and in (d) for $R_{NL}$. I indicates the current injection-collection, V the measurement of the voltage drop.

A further indication of transport via edge states at the CNP in the presence of strong $B_\parallel$, supporting the scenario of the F phase, is obtained with non-local measurements. The contacts’ configuration employed is sketched in Figure 1 (d): the voltage probes are placed as far as possible from the source and drain current contacts. In the case of diffusive transport via bulk states,
the non-local signal \( R_{NL} = V/I \) is expected, according to the de van der Pauw formalism [17], to be exponentially small in the separation between the current contacts and the voltage probes. This is the case of the first trace in Figure 1 (b), which corresponds to \( B_{\parallel} = 0, B_{\perp} = B_{tot} = 7 \) T, at which \( R_{CNP}^{NL}/R_{xx}^{CNP} < 10^{-3} \). The application of increasing \( B_{\parallel} \) has again a tremendous effect, with an increase in \( R_{CNP}^{NL} \) of more than one order of magnitude. Note that, on the other hand, \( R_{xx}^{CNP} \) falls drastically: this rules out the possibility of a dominant bulk contribution to \( R_{CNP}^{NL} \). The strong non-local signal at the CNP, together with the contrasting behavior of the four-probe resistance, can be explained only with transport via edge states. The theoretical value, calculated within the same assumption as in the previous paragraph, is \( R_{CNP}^{NL} = \frac{1}{12} \frac{h}{e^2} \): also in this case the data show a non-perfect quantization.

4. Temperature-dependent measurements and identification of the critical point
We measured \( R_{xx}^{CNP} \) at different values of \( T \) and \( B_{tot} \), while keeping \( B_{\perp} \) constant. In Figure 2 (a) we show the data for \( B_{\perp} = 5 \) T; analogous ones were acquired also at \( B_{\perp} = 4, 6 \) and \( 7 \) T. A smooth insulator-to-metal transition is observed, as expected for the CAF-F [7, 9], while a \( T \)-independent crossing point for the isotherms of \( R_{xx}^{CNP} \) separates the two phases, providing a clear experimental identification of the critical point.

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B_{tot}^* = (2.42 \pm 0.21) \times B_{\perp}
\] (1)

Figure 2 (b) shows the values of critical field \( B_{tot}^* \) corresponding to the \( T \)-independent crossing points as a function of \( B_{\perp} \). \( B_{tot}^* \) clearly increases with \( B_{\perp} \) and a linear dependence can be inferred. From a linear fit to the data (see green line in Figure 2 (b)), we obtain

Equation (1) provides a clear experimental determination of the \( B_{\perp} \)-dependence of this critical field. The Zeeman energies at the critical points \( \epsilon_Z^* \) (see right axis in Figure 2 (b)) are consistent with the CAF-F boundary \( \epsilon_Z \approx 0.8 \) meV reported in Ref. [10] for \( B_{\perp} = 1.75 \) T, which was
estimated on the basis of the saturation of $R_{xx}^{CNP}$ rather than at the insulator-to-metal transition as done here. The reliability of our estimation of the critical point is confirmed by the evolution of the energy gap $\Delta$ of the insulating phase as the in-plane field is increased. In Figure 2 (c) we show that $\Delta$ (obtained by fitting the thermally-activated resistance $R_{xx}^{CNP} \propto \exp(\Delta/2k_BT)$ continuously decreases towards $B_{tot} = B^*_\text{tot}$, eventually pointing at vanishing values, perfectly matching the evolution of the energy gap for the edge excitations which is reported in Ref. [9].

5. Conclusions and future developments

The experimental work we performed, which combines non-local and four-probe measurements of the resistance of a high quality sample, together with the $T$-dependence of the latter, consistently confirm the possibility of inducing a topological phase in bilayer graphene with the use of a strong in-plane magnetic field. We provide a clear identification of the critical field for the CAF-F transition as corresponding to the $T$-independent crossing point for the isotherms of $R_{xx}^{CNP}$. In Ref. [11] we proceed further in the analysis of this transition and propose a scaling behavior of $R_{xx}^{CNP}$ with $(B_{tot} - B^*_\text{tot})/T$ in the vicinity of the critical point.

Future work will be mainly centred on detailed non-local measurements with systematic permutation of the contacts’ configuration (see e.g. Ref. [18]): this will allow determining the exact contribution of each edge path and possibly identifying the origin of the non-exact quantization reported here. Top-gated samples will be employed in order to study transmission, reflection and interference effects of the helical edge channels in the presence of a potential barrier, which will be represented by a fully gapped layer polarized region, locally induced by a vertical electric field. An alternative way of probing the CAF-F transition is represented by resistively-detected NMR measurements [19], which can give a direct estimate of the evolution of the spin polarization at the CNP in bilayer graphene.

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