Transition between canted antiferromagnetic and spin-polarized ferromagnetic quantum Hall states in graphene on a ferrimagnetic insulator

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In the quantum Hall regime of graphene, antiferromagnetic and spin-polarized ferromagnetic states at the zeroth Landau level compete, leading to a canted antiferromagnetic state depending on the direction and magnitude of an applied magnetic field. Here, we investigate this transition at 2.7 K in graphene Hall bars that are proximity coupled to the ferrimagnetic insulator Y1Fe5O12. From nonlocal transport measurements, we demonstrate an induced magnetic exchange field in graphene, which lowers the magnetic field required to modulate the magnetic state in graphene. These results show that a magnetic proximity effect in graphene is an important ingredient for the development of two-dimensional materials in which it is desirable for ordered states of matter to be tunable with relatively small applied magnetic fields (>6 T).

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Graphene has two inequivalent Dirac cones in the energy band dispersion, which lead to a set of Landau levels with distinct features over conventional two-dimensional electron gases, e.g., in an applied magnetic field (B), there exists fourfold degenerate symmetry-broken zero-energy Landau levels with filling factors = 0 and ±1 [1–3]. These are gate-voltage tunable and described by spin and valley degeneracy. Electron-electron and electron-phonon interactions break valley symmetry and determine the magnetic order of the v = 0 state. Theory [4–6] and experiment [7–10] indicate that v = 0 is an antiferromagnetic (AF) quantum Hall state [6,9] in which the two sublattice spins of graphene align antiparallel. The Zeeman field associated with an in-plane magnetic field (B||) favors a spin-polarized ferromagnetic (F) state [9] (that can be also found at v = ±1 [7]), but in general the AF and F states compete, leading to a canted antiferromagnetic (CAF) v = 0 state at a half-filled zero-energy Landau level in which the two sublattice spins tilt out of plane. The spin direction in the CAF state depends on the sum of the spin components parallel (preferred by the Zeeman field, responsible for the F state) and perpendicular to B (preferred by the electron-electron Coulomb interactions responsible for the valley anisotropy and leading to the AF state). In the AF state, there are gapped edge modes while the F state supports gapless counterpropagating edge modes [5,7,11]. Therefore, in the CAF state the energy gap of the edge modes is tunable with the direction and magnitude of B [5].

Edge modes associated with CAF and F states have been detected in graphene Hall bars through nonlocal measurements with a transition between F and CAF states occurring around 15–30 T [9]. In the ballistic limit, the nonlocal resistance (Ra) is quantized and dependent on the Hall bar geometry [12]. In the diffusive limit, Ra is not quantized but shows different behaviors with gate voltage (Vg) in the AF, CAF, and F states [Figs. 1(a)–1(c)]. The AF state does not support edge modes, meaning Ra = 0, but the CAF (F) is gapped (gapless) and Ra shows a double peak (single peak) around the Dirac point (Vg).

Transitions between CAF and F (or AF) states could be achieved in lower applied magnetic fields if graphene has an intrinsic magnetic exchange field (Mex). By placing graphene on an insulating magnetic substrate, a hybridization of the π orbitals in graphene with the substrate can theoretically induce a magnetic exchange field of hundreds of tesla [13–18]. The magnitude of the total magnetic field (MT = gB + Mex) applied to graphene is then related to MT = gB + Mex, where g is gyromagnetic ratio and gB is the Zeeman field. Mex is parallel to B. A 14-T magnetic exchange field was recently estimated in graphene on EuS [14] and an anomalous Hall effect in graphene on Y1Fe5O12 (YIG) showed evidence for

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FIG. 1. Energy spectra for the (a) AF, (b) F, and (c) CAF states in graphene, which arise depending on the total magnetic field ($M_T$) applied to graphene and the angle ($\theta$) between $M_T$ and the sublattice spins. Color scale bar shows $-1$ (spin direction antiparallel to $M_T$) to $1$ (spin direction parallel to $M_T$). Top insets in (a)–(c): Schematic diagrams illustrating the sublattice spins in graphene (left) which make an angle $\theta$ with respect to $M_T$ (right). Bottom insets in (a)–(c): $R_{xx}$ vs $V_{CG}$ for AF, F, and CAF states near $V_D$. (a) The AF state forms when $M_T$ is nonzero (sublattice spins are antiparallel). (b) The F state forms when $M_T$ is larger than a critical value (determined by the Coulomb interaction) and the sublattice spins are parallel to $M_T$. (c) The CAF state is a mixture of AF and F states and forms when $M_T$ is nonzero, but smaller than a critical value (sublattice spins are noncollinear to $M_T$).

an induced magnetic exchange field in graphene [19]. In Refs. [14,19], transitions between CAF and F (or AF) states were not investigated.

Here, we report transitions between CAF and F states in hexagonal boron nitride (hBN) covered graphene Hall bars on YIG. These are investigated through gate-voltage-dependent nonlocal transport measurements below 9 K. The magnetic state and energy gap of the edge modes in graphene are tunable by varying the magnitude (>6 T) and direction of an applied magnetic field ($\mathbf{B}$). The tunable energy gap is important from a fundamental viewpoint, as it separates quantum states with distinct magnetic ordering in graphene, and implies potential relevance for applications requiring a tunable band gap, such as photodetectors.

YIG has a Curie temperature of 550 K, a band gap of 2.84 eV, and an electrical resistivity of $10^{12} \, \Omega \, \text{cm}$. Moreover, it is chemically stable in air, which minimizes surface degradation during Hall bar fabrication. Atomically flat (1 1 0) YIG (84-nm-thick) is prepared by pulsed laser deposition onto gadolinium gallium garnet (GGG) [Fig. 2(a) and bottom inset] with a bulk magnetization of 144 emu cm$^{-3}$ [see Fig. S3 in the Supplemental Material (SM) [21]]. Hall bars are fabricated in several steps, in which graphene is exfoliated from graphite and transferred onto YIG. The graphene is covered with a 20- to 50-nm-thick layer of hBN and electron beam lithography defines the Cr/Au side contacts [21]. In this Rapid Communication, we report two hBN/graphene Hall bars on YIG, which show a field-effect mobility ($\mu$) of around 12 000 cm$^2$ V$^{-1}$ s$^{-1}$ (device I) and 10 000 cm$^2$ V$^{-1}$ s$^{-1}$ (device II) at 9 K. The control Hall bar of hBN/graphene/AIO$_3$/YIG ($\mu \approx 15 000$ cm$^2$ V$^{-1}$ s$^{-1}$ at 9 K) is investigated in which graphene is decoupled from YIG with a 6-nm-thick AIO$_3$ layer. Raman spectroscopy is performed on the graphene prior to and following transfer onto YIG or AIO$_3$ [top inset in Fig. 2(a)] and shows no evidence for defects in graphene.

Figure 2(b) (left inset) shows a representative hBN/graphene Hall bar on YIG prior to top-gate electrode deposition. Resistance is measured using lock-in amplifiers [21]. For local transport, $I_{9,10}$ indicates current flowing between contacts 9 and 10 and a local voltage $V_{3,5}$ is measured between contacts 3 and 5, giving $R_{xx} = V_{3,5}/I_{9,10}$. The nonlocal voltage...
is probed away from the current path (e.g., \( R_{\text{nl}} = R_{34,56} = V_{5,6}/I_{3,4} \)).

We first discuss the transport properties in zero magnetic field for device I at 9 K. Figure 2(b) shows a peak in \( R_{\text{nl}} \) at \( V_D \). By normalizing \( R_{\text{xx}} \) and \( R_{\text{nl}} \) to their maximum values at \( V_D \) (\( R_{\text{xx},D} \) or \( R_{\text{nl},D} \)), we see that \( R_{\text{xx}}/R_{\text{nl},D} \) is an order of magnitude smaller than \( R_{\text{xx}}/R_{\text{xx},D} \) and the peak in \( R_{\text{nl}} \) is sharper than \( R_{\text{xx}} \) [right inset in Fig. 2(b)]. The peak in \( R_{\text{nl}} \) (≈380 Ω) at \( V_D \) may indicate a contribution from the spin Hall [22] or Zeeman spin Hall effects [14]. However, \( R_{\text{xx}} \) shows a negative magnetoresistance (weak localization) [see Fig. S5(b) in SM [21]] at 2.7 K, suggesting that Rashba spin-orbit coupling is not strong at the graphene/YIG interface, and meaning that the spin Hall effect is unlikely to dominate \( R_{\text{nl}} \). The YIG has a small remanent out-of-plane magnetic moment [see Fig. S3(d) in SM [21]] which may support the Zeeman spin Hall effect. We note that ohmic, Joule heating, and Ettingshausen contributions to \( R_{\text{nl}} \) are negligible [21]. Equivalent measurements on an hBN/graphene/AIO\(_x\)/YIG control Hall bar [Fig. 2(c)] show a reduced \( R_{\text{nl}} \) at \( V_D \) of around 65 Ω at 9 K, which is dominated by the ohmic effect. This suggests that in zero magnetic field, \( R_{\text{nl},D} \) of device I is due to a coupling between graphene and YIG.

In Fig. 3(a) we show gate-voltage-dependent Shubnikov–de Haas oscillations in \( R_{\text{xx}} \) and \( R_{\text{nl}} \) (normalized to values at \( V_D \)) for device I with an out-of-plane magnetic field of 2.5 T. The ratios \( R_{\text{xx}}/R_{\text{nl},D} \) and \( R_{\text{nl}}/R_{\text{xx},D} \) show different trends with gate voltage with \( R_{\text{nl}} \) decreasing faster than \( R_{\text{xx}} \). Furthermore, the value of \( R_{\text{nl}} \) is a factor of 50 larger than in the hBN/graphene/AIO\(_x\)/YIG control Hall bar [Fig. 3(b)]. These observations, in conjunction with the fact that the Onsager relation is obeyed in device I, suggest that a coupling between graphene and YIG reduces \( B_{\perp} \) to achieve the \( v = 0 \) state.

For large \( B_{\perp} \), quantum Hall plateaus at \( v = 0 \) and \( ±1 \) may become visible. The \( v = 0 \) state at the half-filled zero Landau level should show a minimum in longitudinal conductance (\( \sigma_{xx} \)) while the other filling factors at a quarter and three-quarter occupancy are at a maximum. In Figs. 4(a)–4(d) we show the dependence of \( \sigma_{xx} \) on gate voltage for increasing values of \( B_{\perp} \). In 4 T a minimum in \( \sigma_{xx} \) is visible at \( V_D \) and approaches zero in 12 T. Over the same magnetic field range at \( V_D \), \( \rho_{\text{xx},D} \) rapidly increases [inset of Fig. 4(d)], indicating a transition to a gapped bulk state. Simultaneously, the Hall conductance (\( \sigma_{xy} \)) tends to be a plateau establishing the \( v = 0 \) state. We note that equivalent measurements on the hBN/graphene/AIO\(_x\)/YIG control Hall bar do not show these trends [21], indicating that a coupling between graphene and YIG reduces \( B_{\perp} \) to achieve the \( v = 0 \) state.

**FIG. 3.** (a) Gate-voltage dependence of \( R_{\text{xx}}/R_{\text{xx},D} \) and \( R_{\text{nl}}/R_{\text{nl},D} \) with \( B_{\perp} = 2.5 \text{T} \). (b) \( R_{\text{nl},D} \) with \( B_{\perp} \) for device I compared to the hBN/graphene/AIO\(_x\)/YIG control Hall bar. (c) \( R_{\text{nl},D} \) vs \( B_{\perp} \) for reverse electrical connections showing that the Onsager relation is obeyed in device I. (d) Landau level fan diagram where the dashed lines are calculated fitting results. Filling factors are shown beside the dashed lines. All data are recorded at 9 K.
The $v = 0$ state in Fig. 4 could be a F or a CAF state. These are distinguishable from the gate-voltage dependence of $R_{n\ell}$ with $B_{\perp}$ as shown in Figs. 5(a)–5(c) for device I and Figs. 5(d)–5(f) for device II; the transition from a single to a double peak in $R_{n\ell}$ with gate voltage suggests that the $v = 0$ state is associated with a transition from the F to CAF state. Although consistent with theory [5], the transition occurs in graphene at lower values of $B_{\perp}$ than without YIG ($> 15$ T in Ref. [9]). Furthermore, the decrease in $R_{n\ell}$ at $V_D$ with increasing $B_{\perp}$ suggests an increase in the edge gap and the angle between $M_T$ and the sublattice spins [21]. This angle increases with $B_{\perp}$ because the valley anisotropy energy (which resulted from electron-electron interactions, which leads to an AF state) increases faster than the Zeeman energy, as discussed in SM [21]. To test this hypothesis, we rotated device II from $\alpha = 90^\circ$ to $\alpha = 0^\circ$ using magnetic fields of 2–12 T, where $\alpha$ is the angle between the Hall bar surface and $B$ [Fig. 5(g)]. This rotation partially (6 T $< ||B|| < 12$ T) or fully ($||B|| = 12$ T) transforms the CAF to F state. As the fixed $B$ rotates in plane, $B_{\perp}$ decreases and the sublattice spins align to $M_T$, reducing the edge gap and increasing $R_{n\ell}$.

Transitions between CAF and F states were investigated in Ref. [9] using hBN/graphene Hall bars with rotating $B$ up to 35 T at 300 mK. In that work the graphene was not in contact with a magnetic substrate, meaning $||M_{\text{ex}}|| = 0$ and thus there is only a Zeeman field. By extracting the average values of $M_T$ vs $B_{\perp}$ in Ref. [9], we calculate a phase transition line of $M_T \approx 9.9 B_{\perp} + 4.9$ which separates the CAF and F states as shown in Fig. 5(h) and explained in SM [21]. For devices I and II [Figs. 5(a)–5(f)], transitions between the CAF and F states
FIG. 5. (a)–(f) Gate-voltage dependence of $R_{nl}$ for different values of $B_\perp$ (labeled) for (a)–(c) device I and (d)–(f) device II. (g) $R_{nl,D}$ vs $\alpha$ with $B_\perp$ from 2 to 12 T for device II. Dashed lines are a guide to the eye. A $\pm 5^\circ$ operational error due to manual rotation of the sample holder leads to the small asymmetry in $R_{nl,D}$ at $\alpha = 60^\circ$ and 120°. (h) Magnetic phase diagram ($M_T$ vs $B_\perp$) for graphene in which the solid (red) line $M_T \approx 9.9 B_\perp + 4.9$ is calculated from Ref. [9] using the extracted data in red as explained in the main text and SM [21]. The blue data represent the estimated phases for devices I and II with $B_\perp$ only. For small $B_\perp$, the quantum Hall state is not well developed. By increasing $B_\perp$, there exists a transition between the F and CAF state. For reasonably small $B_\perp$ and large $M_T$, the F state is realized, whereas by increasing the ratio of $B_\perp/M_T$, the CAF state becomes energetically favored. All data are recorded at 2.7 K except the data from Ref. [9], which are at 300 mK.

In conclusion, we have demonstrated that by proximity inducing a magnetic exchange field in graphene on a ferromagnetic substrate, transitions between CAF and F states can be achieved with relatively low applied magnetic fields ($> 6$ T) at 2.7 K. This achievement is important for the development of two-dimensional materials with magnetic-field-tunable ordered states of matter.

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