The quantum Hall effect near the charge neutrality point in bilayer graphene is investigated in high magnetic fields of up to 35 T using electronic transport measurements. In the high field regime, the eight-fold degeneracy in the zero energy Landau level is completely lifted, exhibiting new quantum Hall states corresponding filling factors $\nu = 0, 1, 2, & 3$. Measurements of the activation energy gap in tilted magnetic fields suggest that the Landau level splitting at the newly formed $\nu = 1, 2, & 3$ filling factors are independent of spin, consistent with the formation of a quantum Hall ferromagnet. In addition, measurements taken at the $\nu = 0$ charge neutral point show that, similar to single layer graphene, the bilayer becomes insulating at high fields.

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The unique chiral nature of the carrier dynamics in graphene results in a novel integer quantum Hall (QH) effect that distinguishes these atomically thin graphitic materials from conventional 2-dimensional systems. The chiral nature of carriers in single layer graphene (SLG) and bilayer graphene (BLG) result in unevenly spaced Landau levels (LL) including a distinctive level located precisely at the particle-hole degenerate zero energy. While the energy spacing of SLG LLs scales as the square root of the LL index and the square root of the field $B$, BLG levels have energies given by

$$\epsilon_n^\pm = \pm \hbar \omega \sqrt{n(n-1)},$$

where $\omega = eB/m$ is the cyclotron frequency with the BLG band mass $m \approx 0.05 m_e$, and $n$ is a non-negative integer representing Landau orbit index in each layer. While the valley and spin degrees of freedom lead to a 4-fold degeneracy in graphene LLs, the additional orbital degeneracy for the $n = 0 \& 1$ indices in BLG result in a 8-fold degenerate, zero energy level that is unprecedented in LL physics.

At high fields, the decreasing radius of the cyclotron orbit gives rise to increasing electron-electron interactions which can perturb the degenerate LLs. Experiments on SLG have demonstrated this high-field symmetry breaking through the appearance of new QH states at the filling factor sequences $\nu = 0, \pm 1, \pm 4$ and $n$ is a non-negative integer representing Landau orbit index in each layer. The precise nature of the field-dependent mechanisms that lift the degeneracies is still under experimental and theoretical debate. In particular, it has recently been observed that in SLG the $\nu = 0$ QH state becomes increasingly insulating at higher fields. Similar to SLG, the enhanced interactions under high magnetic field is expected to lift the 8-fold zero energy LL degeneracy in BLG. A number of theoretical predictions involving unusual collective excitations have been proposed to occur in this particle-hole symmetric LL as its degeneracy is broken, but the experimental observation of this broken symmetry is yet to be observed.

In this Letter we present transport measurements demonstrating the hierarchy of the splittings for the zero-energy LL in bilayer graphene as the external magnetic field is increased. Activation energy measurements of these splittings demonstrate energy spacing smaller than the bare electron-electron interaction energy and Zeeman energy. Tilted field measurements indicate that the degeneracy breaking at filling factors above the charge neutral point are independent of spin, consistent with the formation of a quantum Hall ferromagnet. While a quantum Hall plateau in the transverse resistance is observed at the charge neutral point, the longitudinal resistance at this point is seen to diverge to an insulating state with increasing field.

The graphene samples used in this work are deposited on SiO$_2$(300 nm)/Si substrates by mechanical exfoliation techniques from bulk single crystals. The number of graphene layers are identified by optical contrast, cross-checked using Raman spectroscopy and measurements of the high-field LL spectrum. Au/Cr electrodes for the graphene are fabricated by conventional electron beam lithography followed by metal evaporation. A gate voltage $V_g$ is applied to the degenerately doped Si substrate to control the carrier density $n_e$ according to the relation $n_e = C_g(V_g - V_D)/e$, where the areal gate capacitance is $C_g = 7.1 \times 10^{10} \text{e/V} \cdot \text{cm}^2$ and $V_D$ is the gate voltage corresponding the charge neutrality point. The lower inset of Fig. 1a shows an optical microscope image of a typical BLG device used in this study. This device has a mobility as high as $\sim 1 \times 10^4 \text{cm}^2/\text{V} \cdot \text{s}$ measured at the carrier density $n_h = 4 \times 10^{12} \text{cm}^{-2}$. Since the electrodes of this device are configured in non-ideal Hall bar geometry, the magnetoresistance $R_{xx}$ and Hall resistance $R_{xy}$ were obtained following the van der Pauw method with symmetric ($R_{xx}$) and anti-symmetric ($R_{xy}$) averaging over data from positive and negative magnetic fields. The Hall conductivity $\sigma_{xy}$ is then computed from $R_{xx}$.
QH states corresponding to $\nu = 0$ and $\nu = 2$ states start to develop at 15 T and are fully evolved by 20 T; while the $\nu = 1$ and $\nu = 3$ states start to develop at 20 T and are fully evolved by 25 T. This hierarchical appearance of the new QH states is in accordance with the sequential symmetry breaking of the zero energy LL degeneracy as depicted in Fig. 1(b) and suggests that different symmetry-breaking process are relevant as $B$ increases.

Mechanisms for breaking the LL degeneracy include disorder, lattice strain, and charged impurities. The magnitude of the last of these can be estimated by assuming that the offset of the Dirac point gate voltage $V_g$ from zero energy reflects a bilayer capacitively charged across the $d = 0.34\, \text{nm}$ layer spacing. From this model our measured $|V_D| \sim 2\, \text{V}$ corresponds to a maximum charging energy of $\sim 2\, \text{meV}$, small than, but of the same order as the high-field ($B \gtrsim 20\, \text{T}$) bare Zeeman energy. In addition to the above mentioned graphene impurities, there are two field-dependent factors that can lead to the lifting of the eight degeneracies of this LL: Zeeman splitting of spin in magnetic field and electron-electron interactions that increase with field as the radius of the cyclotron orbits decreases. Zeeman splitting is given by $\Delta_z = g\mu_B B$, where $\mu_B$ is the Bohr magneton, and $g$ is the gyromagnetic factor for the carriers in BLG. The Coulomb interactions between electrons are given by $e^2/\epsilon \ell_B$, where $\epsilon \sim 4$ is the dielectric constant for graphene and $\ell_B = \sqrt{\hbar/eB}$ is the magnetic length. In order to produce full 8-fold symmetry breaking, these mechanisms must lift not only the spin and valley degeneracies, but the $n = 0$ and 1 LL
shown in Fig. 1(b), the mechanism underlying the increasingly insulating behavior of the bilayer at the valley or orbital degeneracy of the zero energy LL. Barlas et al. have suggested that the exchange term from Coulomb interactions between electrons in these degenerate orbital states leads to a QH ferromagnetic state where the 8-fold symmetry is lifted by this exchange interaction. A Hund rule-like hierarchical symmetry breaking in the zero energy LL is predicted with this exchange-enhanced Zeeman splitting followed by the spin-independent valley and orbital splitting [3].

In order to test out this hierarchical degenerate lifting and the role of electron-electron interactions, we first note that sublattice or interlayer splittings that are associated solely with the Coulomb interactions between electrons localized in LL orbits should depend only on the normal magnetic field ($B_\perp$) but not the in-plane field ($B_\parallel$). Whereas spin-splitting includes a Zeeman component determined by the total field $B_{\text{tot}} = \sqrt{B_\parallel^2 + B_\perp^2}$. Experimentally, this idea can be tested out by measuring the samples in a series of tilted fields where we can examine the $R_{xx}$ minima in different $B_\parallel$ and $B_\perp$ by tuning the tilting angle $\theta$ and $B_{\text{tot}}$. Fig. 2 displays the change of $R_{xx}$ as a function of the gate voltage normalized by $B_\perp = B_{\text{tot}} \cos \theta$, in two different experimental conditions.

In Fig. 2(a), we first fix $B_\parallel = 20$ T and then vary $B_\parallel$. For $\nu = 2,3$ QH states, $R^{\text{min}}_{xx}$, the minima of $R_{xx}$ corresponding to the QH zero magnetoresistance, display less than ~10% of variation, indicating no significant changes are induced by applying $B_\parallel$. For comparison, in Fig. 2(b), we show $R_{xx}$ as a function of $B_\parallel$ with fixed $\theta = 0$, that is $B_\parallel = 0$ and $B_\perp = B_{\text{tot}}$. In this case, $R^{\text{min}}_{xx}$ for $\nu = 2$ and $3$ increases by ~40% as $B_\parallel$ decreases from 31 T to 20 T, a factor of 4 larger than the change of Fig. 2(a) where $B_{\text{tot}}$ decreases from 31 T to 20 T while $B_\parallel$ was kept to 20 T. The fact that $R^{\text{min}}_{xx}$ for $\nu = 2$ and 3 shows little to no dependence on the in-plane field strongly suggests that both these QH states are from non-spin origin and thus are due to the breaking of the valley or orbital degeneracy of the zero energy LL.

As for the $\nu = 1$ QH state, the experimental data of its tilted field dependence, shown in the insets of Fig. 2, displays an increasing $R^{\text{min}}_{xx}$ with either an increasing in-plane field or a increasing normal field. While this evolution is consistent neither with an electron-electron or spin origin, it is likely that the increasing $R^{\text{min}}_{xx}$ shown in the inset of Fig. 2(a) is due to the proximity of this filling factor to the increasing insulating behavior of the bilayer at the charge neutrality point, discussed below. However, given the hierarchy of the degeneracy breaking, schematically shown in Fig. 1(b), the mechanism underlying the $\nu = 1$ breaking should be the same as that for $\nu = 3$, i.e. not from a spin origin, consistent with theoretical predications of the formation of a QH ferromagnet in BLG at high magnetic fields [3,14].

To further understand the nature of the fully lifted LL degeneracy, we determine the energy of the LL splittings by measuring $R^{\text{min}}_{xx}$ at different temperatures $T$. Fig. 3(a-c) shows log $R^{\text{min}}_{xx}$ versus $1/T$ for the $\nu = 1,2,3$ states. Since $R^{\text{min}}_{xx} \propto \exp [-\Delta E - 2\Gamma / 2k_B T]$, where $\Delta E$ is the energy gap between two subsequent LLs and $\Gamma$ is the LL broadening. The observed Arrhenius behavior in these plots allows us to estimate $\Delta E$ at different fields from the slope of the line fits. Fig. 3(d) displays the field dependence of the activation gap for $\nu = 1,2,3$. Generally, $\Delta E$ increases with increasing $B$ as expected. It is also noted that at given $B$, $\Delta E_{\nu = 2} > \Delta E_{\nu = 3}$, $\Delta E_{\nu = 1}$, indicating that the even $\nu$ states have larger energy than the odd $\nu$ QH states in accordance with the LL symmetry breaking hierarchy of Fig. 1(b). As for the field dependence, we find that a $\sqrt{B}$ fit is better for $\nu = 2$ and $\nu = 3$ states. Attempts to fit the gap evolution linearly to $B$ result in a positive y-axis intercept for the $\nu = 2$ and $\nu = 3$ gaps and a thus a non-physical negative LL broadening $\Gamma$. From the y-intercept of the $\sqrt{B}$ dependence, we can extract a physically reasonable LL broadening of $\Gamma / k_B = 1.4$ and 3.7 K for the $\nu = 2$ and 3 states, respectively [15]. Although the apparent $\sqrt{B}$-dependence provides an independent confirmation of non-spin origin of the QH states $\nu = 1,2,3$, we note that the observed energy scale $\Delta E$ is still too small compared to the Coulomb energy or even the bare Zeeman energy. For $B = 15$ T, the Coulomb energy is 370 K and the Zeeman energy $E_z = 21$ K, both larger than $\Delta E_{\nu = 2} = 8.6$ K at this field. It is possible that large amounts of disorder near the charge neutrality point of BLG are responsible for such a reduced transport gap.

We finally focus our attention on the $\nu = 0$ QH state. As with SLG, the presumed $\nu = 0$ splitting at the charge neutral point is not directly observable as a zero in $R_{xx}$. 

![Fig. 3: (a) Arrhenius plots of $R^{\text{min}}_{xx}$s as a function of $1/T$ at different fields for $\nu = 1$ state, the lines with respective colors are the linear fits to the data points. (b) Arrhenius plot for $\nu = 2$ state. (c) Arrhenius plot for $\nu = 3$ state. (d) Energy gap $\Delta E$ vs magnetic field $B$ for different filling factors $\nu = 1$ (open triangle red), $\nu = 2$ (open circle green), $\nu = 3$ (open square black). The dotted lines are linear fits, whereas the dashed lines are square-root fits.](image-url)
Rather, $R_{xx}$ displays a maximum at this point, whose value increases with increasing $B$. In Fig. 4, we display our measurement of $R_{xx}$ at the charge neutrality point in BLG. The measured resistance shows quasi-exponential growth as $B$ increases, up to $\sim 10$ MΩ at 30 T. To avoid the self-heating of the graphene discussed in Ref. [12] and to measure resistances $> 10$ MΩ we employ a 2-probe AC measurement configuration with a constant voltage bias of 500 $\mu$V, resulting in only $\sim 10$ fW of heating at the highest fields. The contact resistance included in this measurement set-up is relatively small ($\sim 1$ KΩ), but as an additional confirmation that it does not affect the behavior of the measured resistance, we cross-checked the 2-probe measurement using a conventional, current biased, 4-probe measurement at fields low enough that the resistance could be reliably measured with the voltage probes input to an amplifier with 10 MΩ input impedance. The exponentially divergent $R_{xx}$ behavior at high magnetic field is similar to analogous measurements that have been performed on SLG [11, 12], where a field-induced QH insulator has been proposed. We also note that the gate sweep (inset to Fig. 4) displays a growing number of local maximum in $R_{xx}$, presumably due to the inhomogeneous distribution of these insulating states at high magnetic field [14].

In conclusion, we have observed the full degeneracy lifting of the zero energy LL in bilayer graphene. Independent measurements of the longitudinal resistance zeros for the newly observed filling factors as a function of perpendicular field and temperature each indicate that the degeneracy lifting for the $\nu = 1, 2, & 3$ splittings originate from electron-electron interactions. The field dependence of the longitudinal resistance at the $\nu = 0$ charge neutral point reveals insulating behavior similar in character to that of single layer graphene.

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Note added.—During the preparation of this manuscript, we became aware of related work with a similar conclusion from Feldman et al. [20].

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