Odd Integer Quantum Hall States with Interlayer Coherence in Twisted Bilayer Graphene

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ABSTRACT: We report on the quantum Hall effect in two stacked graphene layers rotated by $2\theta$. The tunneling strength among the layers can be varied from very weak to strong via the mechanism of magnetic breakdown when tuning the density. Odd-integer quantum Hall physics is not anticipated in the regime of suppressed tunneling for balanced layer densities, yet it is observed. We interpret this as a signature of Coulomb interaction induced interlayer coherence and Bose–Einstein condensation of excitons that form at half filling of each layer. A density imbalance gives rise to reentrant behavior due to a phase transition from the interlayer coherent state to incompressible behavior caused by simultaneous condensation of both layers in different quantum Hall states. With increasing overall density, magnetic breakdown gains the upper hand. As a consequence of the enhanced interlayer tunneling, the interlayer coherent state and the phase transition vanish.

KEYWORDS: twisted bilayer graphene, quantum Hall effect, Bose–Einstein condensation, interlayer coherence
Fermi surfaces indicates whether the states originate from the Dirac cone of the reference layer, blue — twisted layer. The twist in real space causes a displacement of the Dirac cones of the two layers in reciprocal space (right) by an amount determined by the twist angle \( \theta \). The black and blue large hexagons indicate the first Brillouin zone of the two layers. The energy where both Dirac cones would intersect increases with increasing twist angle. Also shown is a schematic of the reduced Brillouin zone and the notation used for the symmetry points. (b) For a twisted bilayer, there are two bands. Each of them generates closed isoeneric contours around the \( \bar{K} \) and \( \bar{K}' \) symmetry points below the van Hove singularity. The three panels correspond to three different energies. The Fermi contours associated with the first band are plotted with solid lines. Fermi contours that stem from band 2 are plotted with dotted lines. The coloring of the Fermi surfaces indicates whether the states originate from the Dirac cone of the first (black) or second layer (blue).

Figure 1. Band structure of twisted bilayer graphene. (a) Real-space lattice structure of two graphene layers twisted by an angle \( \theta \) (left, black — reference layer, blue — twisted layer). The twist in real space causes a displacement of the Dirac cones of the two layers in reciprocal space (right) by an amount determined by the twist angle \( \theta \). The black and blue large hexagons indicate the first Brillouin zone of the two layers. The energy where both Dirac cones would intersect increases with increasing twist angle. Also shown is a schematic of the reduced Brillouin zone and the notation used for the symmetry points. (b) For a twisted bilayer, there are two bands. Each of them generates closed isoeneric contours around the \( \bar{K} \) and \( \bar{K}' \) symmetry points below the van Hove singularity. The three panels correspond to three different energies. The Fermi contours associated with the first band are plotted with solid lines. Fermi contours that stem from band 2 are plotted with dotted lines. The coloring of the Fermi surfaces indicates whether the states originate from the Dirac cone of the first (black) or second layer (blue).

twist angles that produce an energy dispersion in the reduced Brillouin zone closely resembling that of graphene with a van Hove singularity due to the merger of the displaced Dirac cones of the original layers (see Figure 1a). The singularity occurs at an energy no longer set by the graphene hopping parameter (about 3 eV), but a much lower twist angle controlled energy. Prior to reaching the van Hove singularity, magnetic breakdown due to the uncertainty in reciprocal space in a magnetic field allows tunneling across momentum gaps between the disjoint and closed Fermi surfaces that form in the reduced Brillouin zone (Figure 1b). The original Dirac cone states at the displaced \( K \)-symmetry points give rise to a band with closed Fermi surfaces encircling the \( \bar{K} \) and \( \bar{K}' \) points of the reduced Brillouin zone (solid circles). The coloring refers to the layer the states originate from (black for layer 1 and blue for layer 2). The same holds for the Dirac cone states at the displaced \( K \)-symmetry points, however with an interchanged location in the reduced zone (dotted Fermi surfaces). The momentum gaps at the reduced zone boundary shrink with increasing density (Figure 1b), and the tunneling probability rises. It is this mechanism that accounts for enhanced "hybridization" or "coupling" between the different layer states with increasing density. The system can then no longer be regarded as composed of two separate layers. When the chemical potential crosses the van Hove singularity, the Fermi surfaces of each band merge into a single Fermi surface, effectively removing the layer degree of freedom all together.

At small angles above the magic angle, the chemical potential can easily be lifted up to this energy with conventional gating techniques. Hence, it is possible to explore the regime of essentially decoupled layers at low densities, weakly tunnel coupled layers at intermediate densities and strongly coupled layers when the chemical potential comes closer to the van Hove singularity. All this can be accomplished at the turn of a gate voltage knob. Devices with twist angles between 2° and 3° represent the sweet spot, because the Fermi velocity in the low energy regime is significantly reduced compared to devices with larger twist angles. The enhanced density of states boosts the importance of Coulomb interactions. Moreover, for twist angles exceeding 5° the overlap matrix element or interlayer transitions between eigenstates of the opposite valleys \( K \) and \( K' \), i.e., between the two Fermi surfaces encircling either the \( K \) and \( K' \) points of the reduced Brillouin zone (Figure 1b), grows rapidly, disrupting the separate layer picture.

As opposed to previous studies focusing either on large twist angles or the magic angle, here we address the transport properties of a device with a twist angle of 2° to search for Bose—Einstein condensation related quantum Hall states for the previously mentioned reasons. For this angle, the van Hove singularity energy is only about 25 meV. A device schematic and image are shown in panels a and b of Figure 2. A bottom and top gate enable the application of a displacement electric field \( D \). Further details of the device and gating characteristics are deferred to the Supporting Information. A color map of the longitudinal resistance recorded across the plane spanned by the total density \( n_{\text{tot}} \) and \( D/\varepsilon_{\text{r}} \) is plotted in Figure 2c. Here, \( \varepsilon_{\text{r}} \) is the permittivity of vacuum. Key features are highlighted: the charge neutrality point at \( n_{\text{tot}} = 0 \), and the two satellite peaks signaling full occupation or depletion of the Moiré potential induced miniband centered around zero energy. The latter appear at a hole and electron density of approximately \( 8.5 \times 10^{12} \) cm\(^{-2} \) (section S2, Supporting Information). The density accommodated by the miniband yields the area of the reduced Brillouin zone from which it is possible to determine the twist...
The momentum gaps between the Fermi surface orbits shrink with increasing carrier density. The layer degree of freedom is effectively removed when the chemical potential approaches the van Hove singularity due to the Lifshitz transition from two to one Fermi surface per band. The 8-fold degeneracy is replaced by a 4-fold degeneracy. Experimental manifestations of this can be seen in Figure 3a. The longitudinal resistivity develops in the absence of a displacement field additional minima at $\nu_{\text{tot}} = \pm 24$ and $\pm 32$, marked by blue arrows in Figure 3a. This coincides with anomalous behavior of the Hall conductance due to coexistence of electrons and holes when the Lifshitz transition occurs.\(^{24}\) This transition is stretched on the $n_{\text{tot}}$ abscissa, since the density of states is large at the singularity. When applying a nonzero displacement, these minima become more pronounced in either direction, and no transition occurs. Hence, the displacement field merely improves the resolution of the layer degeneracy removal, and the two layers act as one layer irrespective of the size of the displacement field in the range accessible in our experiment. This can be seen in Figure 3b along the lines of constant $\nu_{\text{tot}} = \pm 24$ and $\pm 32$. This behavior is distinct from that at other, lower filling factors, i.e., at significantly lower total densities, where a multitude of displacement field induced transitions can be observed. We assert that this difference stems from the decoupled or weakly coupled nature of the layers at these lower fillings/densities, as will be discussed in detail below.
The Hall conductance also develops additional features at fillings $\nu_{\text{tot}} = \pm 24$ and $\pm 32$; however, the observation of a clear transition from $8e^2/h$ steps to $4e^2/h$ steps is hampered by the overall sign reversal of the Hall conductance due to the conversion of the charge carriers from holes to electrons or vice versa, as the chemical potential crosses the van Hove singularity energy.

Figure 4 shows a color map of the longitudinal resistance as a function of total filling factor and displacement field as well as single line traces of both the longitudinal resistance and Hall singularity energy.

Figure 4. (a) Color map of $R_{xx}$ (left) as a function of the displacement field and the total filling factor for $B = 10$ T. The schematic on the right indicates regions where quantum Hall behavior is observed due the simultaneous formation of an integer quantum Hall state in each of the layers. The integer numbers correspond to the filling factor of the top ($\nu_{\text{top}}$) and bottom ($\nu_{\text{bot}}$) layers. (b) (Left) Longitudinal resistance, $R_{xx}$, and (Right) Hall conductivity, $\sigma_{xy}$, as a function of $\nu_{\text{tot}}$ at 10 T when $D/E_0 = 0$ mV/nm. Gray dashed lines mark symmetry broken states. (c) Traces of the longitudinal resistance as a function of $D/E_0$ for different fixed magnetic fields (different colors). All data were recorded at $T = 1.3$ K.

With vanishing resistance. Examples of this can be seen in Figure 4a and c. At $\nu_{\text{tot}} = 2$, the experiment in panel a matches expectations. For $D = 0$, the QHE develops, since both layers take on filling 1. As the displacement field is changed, the top or bottom layer accumulates more charges until it reaches filling $2$, while the other layer is emptied resulting in reentrant behavior. This in principle can continue further with hole fillings for one layer at even larger $D$ resulting in additional transitions. The schematic on the right in panel a of Figure 4 highlights the integer filling of the top and bottom layer at fixed total integer filling for the observed QH-minima in the experimental data on the left. A similar sequence of transitions between QH states also occurs at $\nu_{\text{tot}} = 1$ (Figure 4a) and can be understood in this picture of decoupled layers. However, contrary to expectation, also for $D = 0$ at $\nu_{\text{tot}} = 1$, the bilayer system turns incompressible (green dot in the schematic in panel a) as seen in the single line traces of $R_{xx}$ and $\sigma_{xy}$ recorded at 10 T in Figure 4b as well as at fixed $\nu_{\text{tot}} = 1$ for different $B$-fields in Figure 4c (highlighted by the star). In large twist angle bilayer graphene, only even integer QH states appear for $D = 0$. Odd integer QH physics only emerges for $D \neq 0$. This unexpected $\nu_{\text{tot}} = 1$ state at $D = 0$ in our sample is well separated from the QH behavior due to condensation of both layers into their own quantum Hall state by resistance peaks that appear on either side when moving away from $D = 0$. The resistance peaks are followed by reentrant quantum Hall behavior, because the top and bottom layer approach fillings $\nu_{\text{top}} = 1$ and $\nu_{\text{bot}} = 0$ or vice versa. All QH states including the $D = 0$ state, and the transitions become more pronounced with increasing $B$-field (Figure 4c). We have focused here on electron occupation only. Hole transport is discussed in the Supporting Information, but it is of lesser quality.

In principle, a $\nu_{\text{tot}} = 1$ QH state with $D/E_0$ close to 0 mV/nm may arise either because of interlayer Coulomb interaction driven Bose–Einstein condensation or because a genuine fractional quantum Hall state forms in each of the layers separately, such as, for instance, at $\nu_{\text{top}} = 1/3$ and $\nu_{\text{bot}} = 2/3$ or $\nu_{\text{top}} = \nu_{\text{bot}} = 1/2$. This fractional quantum Hall scenario to produce a $\nu_{\text{tot}} = 1$ for $D = 0$ can, however, be discarded. Our device shows no hint of fractional QH behavior even at 14 T and 30 mK. A half-filled fractional quantum Hall state in an isolated graphene monolayer has only been observed so far when the valley degeneracy is lifted as a result of symmetry breaking by a Moiré superlattice potential imposed by an adjacent hBN film or at magnetic fields exceeding 30 T. Neither condition, however, applies here. Hence, we conclude that the interlayer Coulomb interaction plays a crucial role in reestablishing interlayer coherence in the twisted bilayer. Without interactions, it is absent in essence because of the twist induced moment mismatch of the Dirac cones of the two layers. This is manifested in the absence of odd integer quantum Hall states and odd numerator fractional quantum Hall states in bilayers with large twist angle when $D/E_0 = 0$ mV/nm. Our experimental data demonstrate that interlayer coherence and the accompanied exciton condensation can be restored at balanced densities at lower twist angle, resulting in $\nu_{\text{tot}} = 1$ quantum Hall behavior. Note that with increasing density or field, i.e., as the chemical potential approaches the van Hove singularity, the states of both layers start to hybridize and the system’s behavior should return to that of a single layer. With increasing total density and fields above 12 T, the $\nu_{\text{tot}} = 1$ QH state no longer shows clear reentrant behavior when moving away from $D = 0$. The flanking resistance peaks

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diminish in strength (see Figure S2 of the Supporting Information). In this regime, interlayer coherence no longer needs to be invoked to account for the observed physics.

The quantum Hall effect also appears at \( \nu_{\text{tot}} = 3 \) for balanced layer densities. The emergence of this ground state and its evolution at lower fields are illustrated in Figure 5. The longitudinal resistivity around \( \nu_{\text{tot}} = 3 \) filling as a function of \( D/\epsilon_0 \) is plotted as a color map in panel a for different magnetic field values (3–6 T). At low fields, this \( \nu_{\text{tot}} = 3 \) QH state centered around \( D = 0 \) undergoes again a transition heralded by longitudinal resistance peaks with ascending magnitude of the displacement field followed by reentrant quantum Hall behavior because the two layers condense simultaneously in two different quantum Hall states (here filling 1 or 2 and vice versa for the other layer). At higher field, such transition peaks vanish, completely analogous to the behavior at \( \nu_{\text{tot}} = 1 \). This can be seen in the data recorded at 12 T in Figure S2. The fields at which the layer coherent state associated with exciton condensation disappears is much lower (about three times) for \( \nu_{\text{tot}} = 3 \) than for \( \nu_{\text{tot}} = 1 \). This, however, should not come as a surprise, as this corresponds to about the same total density where single layer behavior is recovered.

In summary, the peculiar dispersion of twisted bilayer graphene consisting of two displaced Dirac cones, that are each composed of states belonging to one of the constituent layers and that hybridize at higher energy due to magnetic breakdown, offers the unique opportunity to modify the effective interlayer coupling strength or tunneling from very weak to strong simply via the mechanism of magnetic breakdown when tuning the density. The layer degree of freedom is effectively removed as the chemical potential approaches the van Hove singularity. These properties have been exploited here to investigate the appearance and evolution of odd integer quantum Hall states with layer tunneling strength. In the regime of low density when the chemical potential is far away from the van Hove singularity and the layers are weakly coupled, the odd integer quantum Hall effect is observed and attributed to interlayer coherence and the formation of a Bose–Einstein condensate of excitons formed by holes and electrons in half filled Landau levels of the two layers. As the chemical potential is raised and the states of both layers hybridize, conventional single layer quantum Hall physics is restored instead.

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nanolett.1c00360.

Sample fabrication and gating details, Top- and back-gate voltage dependence of the longitudinal resistance, Weakening of the reentrant quantum Hall behavior at \( \nu_{\text{tot}} = 1 \), Effect of the graphene electrode with asymmetric gate tuning, Magnetic breakdown in a bilayer with a 2° twist angle (PDF)

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Notes

The authors declare no competing financial interest.

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