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Emergent Dirac gullies and gully-symmetry breaking quantum Hall states in ABA trilayer graphene

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In graphene multilayers, strong trigonal warping of the electronic band structure leads to a complex evolution of Fermi surface topology within the low energy valleys located at the corners of the hexagonal Brillouin zone[1, 2]. The comparatively small energy scales characterizing the underlying interlayer hopping processes (~ 100 meV) renders these transitions accessible via electrostatic gating, providing a highly tunable platform for engineering both zero- and high magnetic field electronic structure. Of particular interest is the possibility to use band structure engineering to create novel manifolds of degenerate Landau levels (LLs), where enhanced electron-electron interaction effects can lead to novel correlated ground states. However, such control comes at the cost of requiring high sample quality to avoid smearing the subtle electronic features.

In this Letter we report magnetocapacitance measurements of exceptionally high quality Bernal stacked trilayer graphene devices. At zero applied magnetic field, we observe a number of electron density- and electrical displacement-tuned features in the electronic compressibility associated with changes in Fermi surface topology. At high displacement field and low density, strong trigonal warping gives rise to three new emergent Dirac cones in each valley, which we term ‘gullies.’ The gullies are centered around the corners of hexagonal Brillouin zone and related by three-fold rotation symmetry. At low magnetic fields of \( B = 1.25 \) T, the gullies manifest as a change in the degeneracy of the Landau levels from two to three. Weak incompressible states are also observed at integer filling within these triplets Landau levels, which a Hartree-Fock analysis indicates are associated with Coulomb-driven nematic phases that spontaneously break rotation symmetry.

To access the high mobility, high-\( D \) regime, we study ABA trilayer flakes encapsulated in hexagonal boron nitride dielectric layers and single-crystal graphite gates[22] (Fig. 1d). We use few-layer graphite to contact the trilayer, allowing us to vary both the total charge density and displacement field \( \vec{D} \) across the trilayer (Fig. 1e). We measure the penetration field capacitance \( C_P \), defined as the capacitance between top and bottom gate with the graphene layer held at constant potential. The finite density of states \( \partial n/\partial \mu \) of the trilayer partially screens the electric field between the top and bottom gate, reducing the measured \( C_P \) so that (for top- and bottom gates with geometric capacitance \( c \)) \( C_P = c^2/(2c + \partial n/\partial \mu) \propto (\partial n/\partial \mu)^{-1} \) for \( \partial n/\partial \mu \gg c \). Changes in \( C_P \) are thus associated with changes in the Fermi surface size or topology.

Fig. 1f shows \( C_P \) measured at \( B = 0 \) as a function of \( D \) and electron density \( n \). A variety of \( n \) and \( D \)-tuned discontinuities are readily visible and indicated in the Figure with numeric labels (1)-(9). These include a sharp \( C_P \) maximum at charge neutrality for both positive and negative \( D \) (1); two elevated \( C_P \) features with parabolic boundaries at negative and positive \( n \) (2-3), two low-\( C_P \) regions with triangular boundary within the parabolic regions (4-5), a ‘wing’-shaped high \( C_P \) region both above and below charge neutrality (6-7), and a narrow elevated \( C_P \) region that runs parallel to the parabolic feature for negative \( n \) bounded by contours (8-9). Some of the capacitance features can be associated with the single-particle band-structure by inspection. For example, (1) is consistent with the small band gap or linear band crossing expected at
Fig. 1. Trilayer graphene band structure and penetration field capacitance measurements at $B = 0$. a. Lattice structure of ABA trilayer graphene with hopping parameters identified. In addition to the $\gamma_i$, the electronic structure is determined by the interlayer potentials $\Delta_1 \propto D$ and the relative potential of the inner layer with respect to the outer layers, $\Delta_2$. b. Electronic band structure of trilayer graphene in the absence of an applied displacement field. The linear monolayer-like and parabolic bilayer-like bands are labeled. The momentum is relative to the $K$ point in the $k_x \parallel \Gamma - K$ direction. c. Band structure evolution under applied electric field. For a wide range of electric fields, the low energy structure is described by three isolated Dirac cones slightly displaced from the $K(K')$ points. d. False color electron micrograph of the measured trilayer graphene device. The active region is indicated in cyan. e. Device schematic: trilayer graphene encapsulated in $\sim 20$ nm BN with few-layer graphite top and bottom gates. Independent contacts to the gates and graphene layer allow independent control of charge density $n = c_i V_i + c_6 V_6$ and displacement electric field $D = \varepsilon_{BN} (V_1/d_1 - V_6/d_6)$, where $c_{BN} \approx 3$ and $d_1$ is the distance to the gate. f. Penetration field capacitance $C_P$ at $B = 0$ T and $T \approx 50$ mK as a function of $n$ and $D$. $D$ breaks the mirror symmetry of the ABA-stacked trilayer graphene and induces an on-site energy difference $\Delta_1$ between the outer layers. Main features visible in the experimental data are indicated by dashed lines and numerals. The $D < 0$ region is shaded to increase the visibility of the features. Data is plotted on a saturated color scale (see Fig. S5).

To understand the remaining observed compressibility features, we perform tight binding simulations of the trilayer graphene band structure. Energy eigenvalues are computed using a 6-band tight binding model (see Supplementary information). Hopping between different atoms within the unit cell is parameterized by six tight binding parameters $\gamma_i$, $i = 1, \ldots, 6$, one on-site energy $\delta$, and two energy asymmetries $\Delta_1$ and $\Delta_2$. $\Delta_1$ describes the potential difference between the top and bottom layers and is most directly tuned by the strength of an externally applied polarizing electric field $D$. $\Delta_2$ measures the potential imbalance between the central layer and the two outer layers, and screening effects within the trilayer.

Figure 2a shows the calculated inverse compressibility within this model, as a function of the carrier density and $\Delta_1 \propto |D|$. Both the geometric and parasitic capacitances within the device influence the mapping of $\partial n / \partial \mu \leftrightarrow C_P$ between calculated compressibility and measured data. Moreover, interactions likely renormalize the compressibility particularly when it is high. We thus restrict ourselves to qualitative comparisons of the magnitude of the signals, and plot both in arbitrary units. We do, however, achieve quantitative agreement between data and simulation for the position of extrema and discontinuities for parameters $\gamma_0 = 3.1$, $\gamma_1 = .38$, $\gamma_2 = -0.021(5)$, $\gamma_3 = 0.29$, $\gamma_4 = 0.141(40)$, $\gamma_5 = 0.050(5)$, $\delta = 0.0355(45)$, and $\Delta_2 = 0.0035$, where all energies are expressed in eV. Notably, the model succeeds in matching the experimentally observed features only for an exceptionally narrow range of parameters, providing tighter constraints on $\gamma_i$ and $\Delta_i$ than previously achieved using only LL coincidences[13, 17, 25]. In addition to the parameters $\gamma_i$ and $\Delta_2$, a single scale factor $\alpha = .165$ e$\cdot$nm is chosen so that $\Delta_1 = \alpha \cdot D$. $\alpha$ describes dielectric screening of the perpendicular electric field by the trilayer, implying an effective $\varepsilon_{\text{TLG}}^* \approx 4$ for the trilayer itself (see supplementary information).

The agreement between theory and experiment allows us to understand the connection between the observed compressibility features and the nature of the Fermi contours.Fig. 2b shows calculated Fermi surface contours in 11 distinct regions throughout the experimentally accessed parameter regime. Regions (i) and (xi), for example, are distinguished by the existence of a second, independent Fermi surface arising from the second electron- or hole-subband, respectively, as intuited above. All other regions are separated by Lifshitz transitions and distinguished by differences in Fermi surface topology within a single electron- or hole-band. We note that signatures of Lifshitz transitions were recently found in tetralayer graphene[2] at zero magnetic field, but no direct compressibility measurements of Lifshitz transitions have been reported.
With the exception of regions iii-iv, all of the regions are bounded by experimentally observed features described in Fig. 1. We note that features characterized by a diverging density of states, such as the iii-iv boundary, only weakly modify the measured capacitance and are barely discernible even in Fig. 1f.

Fig. 2c-d shows comparisons of traces from the measured capacitance and the numerically calculated inverse compressibility at \( n = -1.0 \times 10^{12} \text{cm}^{-2} \). Both data and simulation show matching discontinuities associated with the band edge of the second hole subband (i.e., the xi-x transition) as well as the nucleation of new electron pockets within the main hole-pocket (x-i-x transition). Of particular interest is the regime of low \( n \) and large \( D \), where the gully Dirac points are predicted[6].

In addition to its thermodynamic signatures at \( B = 0 \), the emergence of isolated Dirac cones can be expected to lead to new transport, optical, and thermodynamic phenomenology at finite magnetic fields. In monolayer graphene, for example, the two inequivalent valleys lead to four-fold internal degeneracy of the LLs, with an additional factor of two arising from electron spin. The observation of four-fold degeneracy was a critical feature of the first experimental demonstrations of the Dirac spectrum in monolayer graphene[26, 27].

The gully Dirac cones similarly manifest as increased LL degeneracy. Figure 3a shows \( C_P \) data measured at \( B=1.25 \text{T} \) alongside the results of diagonalizing the trilayer Hamiltonian in the presence of a magnetic field (simulations ignore spin splitting; see supplementary information). Larger energy gaps manifest as prominent peaks in \( C_P \) at filling factors \( \nu = \frac{eBn}{\hbar} \), spaced by integer multiples of \( g \), the internal LL degeneracy. Near \( D = 0 \), we observe the strongest capacitance peaks spaced by \( \Delta \nu = 2 \), in agreement with the two-fold valley degeneracy (\( g = 2 \)) but lifted spin degeneracy (Fig. 3b, top). In contrast at large displacement fields (\( D > 0.7 \text{V/nm} \)) and near charge neutrality—i.e., in the regime of the Dirac gullies—this behavior changes, with the most prominent gaps spaced by \( \Delta \nu = 3 \) for \(-12 < \nu < 12 \) (see Fig. 3b, bottom). The calculated single particle energy spectrum (Fig 3c) shows that displacement field leads to the formation of four triplets of LLs per spin projection (labeled T1, T2, T3, and T4); within each triplet, three LLs intertwine into a single three-fold quasi-degenerate band consistent with the observed LL degeneracy. We note that triplet LLs are a generic feature of trigonally warped multilayer band structures, and evidence for three-fold degenerate LLs has previously been reported in suspended bilayer graphene samples[28].

While the observation of triplet LLs is consistent with expectations from our single-particle model, close examination of high \( D \) data reveals departures from the noninteracting picture. In particular, we observe \( C_P \) peaks at all integer filling factors \(-6 < \nu < 12 \), corresponding to the dashed region of Fig. 3a (see also Fig. S6), including weak peaks at \( (\nu \mod 3) \neq 0 \). These gaps persist without closing over the whole range of \( D > 0.7 \text{V/nm} \). This is qualitatively inconsistent with the single particle spectrum, which predicts
that within each triplet (T1...T4 in Fig. 3c) the single particle eigenstates evolve via a series of crossings with increasing $\Delta_1$ (Fig. 3d). One thus expects these anomalous gaps to undergo repeated closings, in contrast to their observed persistence.

The failure of the single-particle picture is not surprising. The estimated bandwidth of each triplet (Fig. 3d), $\delta \varepsilon < 0.5$ meV, is smaller than the scale of the Coulomb interactions, $E_C = e^2/\epsilon \ell_B \approx 10$ meV at $B = 1.25$ T (here $e$ is the elementary charge, $\epsilon = 6.6$ the in-plane dielectric constant of hBN[29], and $\ell_B = \sqrt{\hbar/eB}$ the magnetic length). Taking these interactions into account, the individual LLs within the triplet are effectively degenerate; the ground state at integer filling must result from minimizing repulsive interactions and is likely to result in a gapped, symmetry breaking quantum Hall ferromagnetic state.

We investigate this quantitatively using a variational Hartree-Fock analysis (see supplementary information) of the ground state when only one out of 3 LLs within a single spin branch of triplet T2 is filled (1/3 filling). The three insets to Fig. 3d show real space probability distributions for coherent states constructed for each of the three components of T2. Absent interactions, the ground state at 1/3 filling consists of the lower energy component of T2 for a given value of $B$ and $\Delta_1$, and preserves rotation symmetry. In contrast, the Hartree-Fock ground state (Fig. 3e) spontaneously breaks the $C_3$ symmetry—it is a gully nematic. As long as $\delta \varepsilon \ll E_C$, the gap will be only weakly modulated by $\Delta_1$, making it insensitive to the single-particle level crossings, in agreement with experimental observation.

The nematic ground state is merely one example of a symmetry breaking channel. Intuitively, nematics are favored by interactions when LL wave functions are localized in well separated real-space pockets, as in the case in the highly anisotropic wave functions of Fig. 3e. In a momentum space picture, these pockets are associated with the main Dirac gulies represented in the contours of Fig. 2b vii. In this limit, ABA trilayer triplet LLs resemble the case of the (111) surface of SnTe recently considered theoretically. [30]. Our single-particle calculations suggest that other limiting behaviors can also be realized in ABA trilayer graphene, resulting in qualitatively different ground states. For instance, the triplet states T1 and T4 are considerably less anisotropic, being associated with multiple momentum space pockets close to the $K(K')$ points as in Fig 2b vii. In these triplets, isotropic ground states constructed from a superposition of triplet wavefunctions may be favored. Notably, the relevant anisotropies within each triplet are continuously tunable by external electric and magnetic fields, making ABA trilayer graphene an remarkably versatile platform for exploring correlation effects in unusual quantum Hall ferromagnets. Cataloging the theoretical possibilities, and determining how to distinguish them experimentally, will be the topic of future work.
[1] Edward McCann and Vladimir I. Fal’ko. Landau-Level Degeneracy and Quantum Hall Effect in a Graphite Bilayer. Phys. Rev. Lett., 96(8):086805, March 2006.

[2] Yanneng Shi, Shi Che, Kuan Zhou, Supeng Ge, Ziqi Pi, Timothy Espiritu, Takashi Taniguchi, Kenji Watanabe, Yafis Barlas, Roger Lake, and Chun Ning Lau. Tunable Lifshitz Transitions and Multiband Transport in Tetraylere Graphene. Physical Review Letters, 120(9):096802, February 2018.

[3] Mikito Koshino and Edward McCann. Gate-induced interlayer asymmetry in ABA-stacked trilayer graphene. Phys. Rev. B, 79(12):125443, March 2009.

[4] B. Partoens and F. M. Peeters. From graphene to graphite: Electronic structure around the K point. Physical Review B, 74(7):075404, August 2006.

[5] A. A. Avetisyan, B. Partoens, and F. M. Peeters. Electric-field control of the band gap and Fermi energy in graphene multilayers by top and back gates. Physical Review B, 80(19):195401, November 2009.

[6] Maksym Serbyn and Dmitry A. Abanin. New Dirac points and multiple Landau level crossings in biased trilayer graphene. Physical Review B, 87(11):115422, March 2013.

[7] Takahiro Morimoto and Mikito Koshino. Gate-induced Dirac cones in multilayer graphenes. Physical Review B, 87(8):085424, February 2013.

[8] Inti Sodemann, Zheng Zhu, and Liang Fu. Quantum Hall Ferroelectrics and Nematics in Multivalley Systems. Physical Review X, 7(4):041068, December 2017.

[9] Benjamin E. Feldman, Mallika T. Randeria, András Gycins, Fengcheng Wu, Huiwen Ji, R. J. Cava, Allan H. MacDonald, and Ali Yazdani. Observation of a nematic quantum Hall liquid on the surface of bismuth. Science, 354(6310):316–321, February 1992.

[10] M. F. Craciun, S. Russo, M. Yamamoto, J. B. Oostinga, A. F. Morpurgo, and S. Tarucha. Trilayer graphene is a semimetal with a gate-tunable band overlap. Nature Nanotechnology, 4(6):383–388, June 2009.

[11] A. Kumar, W. Escoffier, J. M. Poumirol, C. Faugeras, D. P. Avrames, M. M. Foger, F. Guinea, S. Roche, M. Goiran, and B. Raquet. Integer Quantum Hall Effect in Trilayer Graphene. Physical Review Letters, 107(12):126806, September 2011.

[12] E. A. Henriksen, D. Nandi, and J. P. Eisenstein. Quantum Hall Effect and Semimetallic Behavior of Dual-Gated ABA-Stacked Trilayer Graphene. Physical Review X, 2(1):011004, January 2012.

[13] Thiti Taychatanapat, Kenji Watanabe, Takashi Taniguchi, and Pablo Jarillo-Herrero. Quantum Hall effect and Landau-level crossing of Dirac fermions in trilayer graphene. Nature Physics, 7(8):621–625, August 2011.

[14] W. Bao, L. Jing, J. Velasco, Y. Lee, G. Liu, D. Tran, B. Standley, M. Aykol, S. B. Cronin, D. Smirnov, M. Koshino, E. McCann, M. Bockrath, and C. N. Lau. Stacking-dependent band gap and quantum transport in trilayer graphene. Nature Physics, 7(12):948–952, December 2011.

[15] Yongjin Lee, Jairo Velasco, David Tran, Fan Zhang, W. Bao, Lei Jing, Kevin Myhro, Dmitry Smirnov, and Chun Ning Lau. Broken Symmetry Quantum Hall States in Dual-Gated ABA Trilayer Graphene. Nano Letters, 13(4):1627–1631, April 2013.

[16] L. C. Campos, A. F. Young, K. Surakhitbovorn, K. Watanabe, T. Taniguchi, and P. Jarillo-Herrero. Quantum and classical confinement of resonant states in a trilayer graphene Fabry-Pérot interferometer. Nature Communications, 3:1239, December 2012.

[17] Yuya Shimazaki, Toru Yoshizawa, Ivan V. Borzenets, Ke Wang, Xiaomeng Liu, Kenji Watanabe, Takashi Taniguchi, Philip Kim, Michihisa Yamamoto, and Seigo Tarucha. Landau level evolution driven by band hybridization in mirror symmetry broken ABA-stacked trilayer graphene. arXiv:1611.02395 [cond-mat], November 2016. arXiv: 1611.02395.

[18] Petr Stepanov, Yafis Barlas, Tim Espiritu, Shi Che, Kenji Watanabe, Takashi Taniguchi, Dmitry Smirnov, and Chun Ning Lau. Tunable Symmetries of Integer and Fractional Quantum Hall Phases in Heterostructures with Multiple Dirac Bands. Physical Review Letters, 117(7):076807, August 2016.

[19] Biswajit Datta, Santanu Dey, Abhishek Samanta, Hitesh Agarwal, Abhinandan Borah, Kenji Watanabe, Takashi Taniguchi, Rajdeep Sensarma, and Mandar M. Deshmukh. Strong electronic interaction and multiple quantum Hall ferromagnetic phases in trilayer graphene. Nature Communications, 8:14518, February 2017.

[20] Mikito Koshino and Edward McCann. Landau level spectra and the quantum Hall effect of multilayer graphene. Physical Review B, 83(16):165443, April 2011.

[21] Shengjun Yuan, Rafael Roldán, and Mikhail I. Katsnelson. Landau level spectrum of ABA- and ABC-stacked trilayer graphene. Physical Review B, 84(12):125455, September 2011.

[22] A. A. Zibrov, C. Kometter, H. Zhou, E. M. Spanton, T. Taniguchi, K. Watanabe, M. P. Zaletel, and A. F. Young. Tunable interacting composite fermion phases in a half-filled bilayer-graphene Landau level. Nature, 549(7672):360–364, September 2017.

[23] J. P. Eisenstein, L. N. Pfeiffer, and K. W. West. Negative compressibility of interacting two-dimensional electron and quasi-particle gases. Phys. Rev. Lett., 68(5):674–677, February 1992.

[24] R. C. Ashoori, H. L. Stormer, J. S. Weiner, L. N. Pfeiffer, S. J. Pearton, K. W. Baldwin, and K. W. West. Single-electron capacitance spectroscopy of discrete quantum levels. Phys. Rev. Lett., 68(20):3088–3091, May 1992.

[25] Leonardo C. Campos, Thiti Taychatanapat, Maksym Serbyn, Kawin Surakhitbovorn, Kenji Watanabe, Takashi Taniguchi, Dmitry A. Abanin, and Pablo Jarillo-Herrero. Landau Level Splittings, Phase Transitions, and Nonuniform Charge Distribution in Trilayer Graphene. Physical Review Letters, 117(6):066601, August 2016.

[26] Yuanbo Zhang, Yan-Wen Tan, Horst L. Stormer, and Philip Kim. Experimental observation of the quantum Hall effect and Berry’s phase in graphene. Nature, 438(7065):201–204, November 2005.

[27] K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, M. I. Katsnelson, I. V. Grigorieva, S. V. Dubonos, and A. A. Firsov. Two-dimensional gas of massless Dirac fermions in graphene. Nature, 438(7065):197–200, November 2005.

[28] Anastasia Varlet, Dominik Bischoff, Pauline Simonet, Kenji Watanabe, Takashi Taniguchi, Thomas Ihn, Klaus Ensslin, Marcin Mucha-Kruczynski, and Vladimir I. Fal’ko. Anomalous Sequence of Quantum Hall Liquids Revealing a Tunable Lifshitz Transition in Bilayer Graphene. Physical Review Letters, 113(11):116602, September 2014.

[29] R. Geick, C. H. Perry, and G. Rupprecht. Normal Modes in Hexagonal Boron Nitride. Physical Review, 146(2):543–547, June 1966.

[30] Xiaol Li, Fan Zhang, and A. H. MacDonald. SU(3) Quantum Hall Ferromagnetism in SrTe. Physical Review Letters, 116(2):026803, January 2016.

[31] See Supplemental Material [url] for additional data and theor etic.
illegal simulations, which includes Refs. [32-39].

[32] M. S. Dresselhaus and G. Dresselhaus. Intercalation compounds of graphite. *Advances in Physics*, 51(1):1–186, January 2002.

[33] C. L. Lu and C. P. Chang and Y. C. Huang and R. B. Chen and M. L. Lin. Influence of an electric field on the optical properties of few-layer graphene with AB stacking. *Physical Review B*, 73(14):144427, April 2006.

[34] F. Guinea, A. H. Castro Neto, and N. M. R. Peres. Electronic states and Landau levels in graphene stacks. *Physical Review B*, 73(24):245426, June 2006.

[35] Hongki Min, Bhagawan Sahu, Sanjay K. Banerjee, and A. H. MacDonald. Ab initio theory of gate induced gaps in graphene bilayers. *Physical Review B*, 75(15):155115, April 2007.

[36] A. Grneis, C. Attaccalite, L. Wirtz, H. Shiozawa, R. Saito, T. Pichler, and A. Rubio. Tight-binding description of the quasiparticle dispersion of graphite and few-layer graphene. *Physical Review B*, 78(20):205425, November 2008.

[37] Biswajit Datta, Hitesh Agarwal, Abhisek Samanta, Amulya Ratnakar, Kenji Watanabe, Takashi Taniguchi, Rajdeep Sen-sarma, and Mandar M. Deshmukh. Landau level diagram and the continuous rotational symmetry breaking in trilayer graphene. *arXiv:1802.05691* [cond-mat], February 2018. arXiv: 1802.05691.

[38] A. H. MacDonald. Influence of Landau-level mixing on the charge-density-wave state of a two-dimensional electron gas in a strong magnetic field. *Physical Review B*, 30(8):4392–4398, October 1984.

[39] Fan Zhang, Dagim Tilahun, and A. H. MacDonald. Hund’s rules for the SN=0S Landau levels of trilayer graphene. *Physical Review B*, 85(16):165139, April 2012.