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Phys. Rev. Lett. **128**, 217701 — Published 23 May 2022
DOI: 10.1103/PhysRevLett.128.217701

Observation of re-entrant correlated insulators and interaction driven Fermi surface reconstructions at one magnetic flux quantum per moiré unit cell in magic-angle twisted bilayer graphene

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The discovery of flat bands with non-trivial band topology in magic angle twisted bilayer graphene (MATBG) has provided a unique platform to study strongly correlated phenomena including superconductivity, correlated insulators, Chern insulators and magnetism. A fundamental feature of the MATBG, so far unexplored, is its high magnetic field Hofstadter spectrum. Here we report on a detailed magneto-transport study of a MATBG device in external magnetic fields of up to B = 31 T, corresponding to one magnetic flux quantum per moiré unit cell Φ_0. At Φ_0, we observe re-entrant correlated insulators at a flat band filling factors of $\nu = +2$ and of $\nu = +3$, and interaction-driven Fermi surface reconstructions at other fillings, which are identified by new sets of Landau levels originating from these. These experimental observations are supplemented by theoretical work that predicts a new set of 8 well-isolated flat bands at Φ_0, of comparable band width, but with different topology than in zero field. Overall, our magneto-transport data reveals a qualitatively new Hofstadter spectrum in MATBG, which arises due to the strong electronic correlations in the re-entrant flat bands.

Two superimposed graphene monolayers, misaligned by a small twist angle $\theta = 1.1^\circ$, produce 8 electronic flat bands near the charge neutrality point (CNP) [1]. These bands host strong electronic interactions and provide an exciting platform to study exotic quantum phenomena, such as correlated insulators [2–5], superconductors [3, 4, 6–8], magnets [3, 9–11], strange metals [12, 13], etc. The moiré super-potential in MATBG creates a new set of renormalized bands in a mini-Brillouin zone, which possess C_{2z}, C_{3z}, C_{2x} rotational symmetries and time reversal symmetry T at zero magnetic field. Non-zero Dirac helicity in MATBG, protected by the $C_{2z}T$ symmetry and the decoupled valleys, gives rise to the non-trivial band topology [14-16]. As the role of the symmetries in the observed quantum phases in MATBG is still a topic of active investigation, discovering novel flat band systems with different inherent symmetries, but similar phenomenology, is a major goal in the field.

Our exact theoretical study of the Bistritzer-MacDonald (BM) Hofstadter spectrum has uncovered a set of low-energy flat bands at one quantum of magnetic flux per moiré unit cell Φ_0[17]. While strong magnetic fields tend to break the bands into fractal Hofstadter sub-bands, here due to the Aharonov-Bohm effect, full density Bloch-like bands re-emerge at Φ_0. The moiré unit cell area of MATBG is almost 3000 times larger than the graphene unit cell; thus, the magnetic field required to reach one full flux quantum is far smaller, close to 30 T, and is within experimental reach [18, 19]. Although previous studies of the electronic properties of graphene/hBN superlattice in the presence of high magnetic fields have provided great insight into the Hofstadter spectrum [20-27], flat electronic bands with strong correlations have never been explored in this light before. Together, these characteristics make MATBG an unprecedented platform to study the Hofstadter spectrum enriched by band topology and strong interactions.

Here, we report on the detailed magneto-transport behavior of a MATBG device with a twist angle $\theta = 1.12° \pm 0.02°$ in the presence of an external perpendicular magnetic field as high as 31 T. We resolve the continuous evolution of the Hofstadter spectrum from zero field to Φ_0, which corresponds to a B-field of $B_0 = 30.5$ T. At B_0 we observe re-entrant correlated insulators at certain integer fillings and interaction-driven Fermi surface reconstructions, evidenced by new sets of Landau levels originating from these. We also study the higher energy passive bands, where we observe a rich interaction-reconstructed Hofstadter spectrum.

Fig. 1a shows the schematic of the device, which comprises a van der Waals heterostructure of graphite/hBN/MATBG/hBN, where the hBN layers were specifically non-aligned with the MATBG. We performed four-terminal longitudinal resistance R_{xx} and Hall resistance R_{xy} measurements where the carrier density n of the band is continuously tuned by a gate voltage V_g. The total carrier density n is normalized by $n_S = 4n_0$, where n_S is the density of the fully filled spin and valley degenerate moiré bands and n_0 is the density per flavor. The filling factor of the carriers per moiré unit cell is defined as $\nu = n/n_0$.

Fig. 1b illustrates the band structure of MATBG with $\theta = 1.12°$ at zero flux and at Φ_0. The zero-flux band structure comprises two connected nearly flat topological bands and higher-energy dispersive bands. An exact study of the BM Hamiltonian in magnetic flux predicts the emergence of a set of low-energy flat bands at Φ_0 but with different symmetry and topology from the zero flux flat bands [17].

Fig. 1d shows the line plots of R_{xx} vs. ν at $B = 0$ T (zero flux) and $B = 30.5$ T (Φ_0) at $T = 40$ mK. The $B = 0$ T trace is dominated by a well-known sequence of resistance peaks [2, 3]. The charge neutrality point (CNP) appears to be a gapless phase since R_{xx} is of the order of 10 kΩ. We observe correlated insulators (CI) at $\nu = \pm 2$, +3, and band insulators (BI) at $\nu = \pm 4$. In addition, we observe a superconducting region near $\nu = -2$ (see Extended data E). In the $B = 30$ T trace, we observe an overall similar picture, which strikingly shows enhanced resistance peaks at $\nu = +2$ and +3: a first indication of re-entrant CIs. Temperature dependence of these states (Extended data D) shows a clear insulating behaviour. Nevertheless, some differences emerge. The CNP is now gapped with a resistance in the order of $R_{xx} \sim 10$MΩ, and the gaps of the BIs at $\nu = \pm 4$ are enhanced. We also do not find signatures of superconductivity. We argue that the experimental parameters to observe SC are much more stringent than the CIs. Having a low critical magnetic field ($B_c \sim 50$ mT) makes this system very challenging to observe SC at Φ_0 flux. Since a tiny twist-angle inhomogeneity of $\Delta\theta = 0.05°$ will give rise to a variation of the full flux value by $\Delta B = 60$ mT, which is enough to fully suppress the SC state. We have discussed this in detail in Extended data E.

To highlight the re-entrant behavior of the $\nu = 2$ CI at Φ_0, we show the evolution of the R_{xx} vs. ν in B-field in the color plot of Fig. 1c. The ν-B phase space appears to be nearly symmetric about the point corresponding to $\nu = 2$ and $B = 15$ T $= 0.5\ \Phi_0$, under an approximate symmetry sending ν to 4-ν and B to Φ_0 -B. At $B = 0$ T we observe a set of Landau levels with LL filling factors $\nu_L = +2$, +4 etc. originating from the CI at $\nu = 2$. The LL with $\nu_L = +2$ was previously interpreted as a correlated Chern insulator (CCI) with a Chern number $C = 2$ [28]. The corresponding R_{xx} and R_{xy} vs. ν line traces for $B =$ 5 T are plotted in Fig. 1f, which show clear quantum Hall signatures of $\nu_L = +2$ with $R_{xx} \sim 0$ Ω and R_{xy} ~ 12.5 kΩ. The strong interaction opens up a gap at $\nu = 2$ and reconstructs the Fermi surface, which results in a degeneracy of 2 by lifting the 4-fold spin-valley degeneracy.

The CI is continuously suppressed with increasing B and vanishes at $B \sim 8$T, where the phase diagram becomes dominated by LLs. Strikingly, above $B > 24$T the $\nu = 2$ CI state reappears and grows continuously stronger upto Φ_0 (see Extended data C). While due to experimental limitations we could not measure our device in B-fields well above Φ_0, by continuity, all the LLs that point away from the CNP above B_0, will point towards the CNP below B_0. Similar to zero flux, we also observe the emergence of a set of LLs from $\nu = 2$ and Φ_0, with $\nu_L = +2$, (+3), +4, (+5). Here the even fillings are more pronounced than the odd ones, as can be seen in Fig. 1e, which shows the quantum Hall traces at $B = 22$ T. In summary, the occurrence of an insulating state which is accompanied by a Fermi surface reconstruction at Φ_0 firmly establishes the existence of a re-entrant $\nu = 2$ CI. Since the most dominant LL here is the $\nu_L = +2$, we also speculate that this LL can be interpreted as a CCI with $C = 2$, in direct analogy to the $B = 0$ T case [28].

We further examine the full ν-B phase space of the flat bands. Figs. 2a and 2b show the respective Landau fan diagrams of R_{xx} and R_{xy} as a function of ν and B, where the observed LLs are schematically laid out in Fig. 2c. At 0 flux we observe a set of LLs with degeneracy 2 originating from $\nu = \pm 2$, fully non-degenerate LLs from the CNP, LLs with $\nu_L = \pm 3$ from $\nu = \pm 1$ and LLs with $\nu_L = \pm 1$ from $\nu = \pm 3$, in agreement with previous studies [28-30, 36].

At Φ_0, we observe a multitude of new sets of LLs emerging from the different integer fillings ν, demonstrating abundant Fermi surface reconstructions. Surprisingly it appears that all the Fermi surfaces have a fully lifted degeneracy, where from $\nu = +1$ we find LLs with the sequence $\nu_L = +1, +2, +3$, from $\nu = \pm 2$ we find LLs with $\nu_L = \pm 2, \pm 3, \pm 4, \pm 5$, and from $\nu = \pm 3$ we find LLs with $\nu_L = \pm 1, \pm 2, \pm 3, \pm 4$. This suggests that for both odd and even integers the spin and valley degeneracies have been lifted at Φ_0 in contrast to zero flux. This can be attributed to the breaking of $C_{2z}T$ symmetry by magnetic flux [18, 31-33] which lifts the degeneracy of the quasiparticles on top of the CIs, although quantitative predictions have not been made so far at odd fillings. Furthermore, we also observe LLs which survive through the full range of magnetic field and connect two different integer fillings at zero flux and at Φ_0. Whereas Ref. [17] uses a strong coupling approach to analyze the ground states at Φ_0, more theoretical work is required to study the interactions within the Hofstadter sub-bands at intermediate flux.

The strong interaction at zero flux spontaneously breaks the $C_{2z}T$ symmetry and can give rise to several Chern bands at different odd integer fillings [17, 31, 34]. At Φ_0, the $C_{2z}T$ symmetry is broken by the magnetic field, leading to different single-particle topology in the flat bands and different degeneracies in the many-body charge excitations atop the CI states [17]. The observed differences in the $B = 0$ T and $B = 30.5$ T LLs reflect the importance of symmetry and topology in determining the many-body phases. The observation of these LLs and the interaction-driven CIs confirms the existence of the (theoretically predicted) flat bands at Φ_0.

The higher energy passive bands in MATBG have larger bandwidth than the flat bands and are amenable to a single-particle Hofstadter analysis. Using the new gauge-invariant formalism of [35], we numerically compute the spectrum to very high accuracy between $\Phi = 0$ and Φ_0. The results are shown in Fig. 3a where the Chern numbers of the largest gaps are computed using Wilson loops and the Streda formula. We emphasize that our calculations are exact within the BM model, and do not rely on $\boldsymbol{k}\cdot\boldsymbol{p}$ approximations near the Fermi surfaces. To access this regime experimentally, we have tuned the carrier density above $\nu > 4$. Fig. 3b shows the color plot of R_{xx} as a function of the normalized magnetic flux and ν. The strongest LLs, observable as lines with slope $1/\nu_L$, are schematically laid out in Fig. 3c with corresponding (ν, ν_L), where we find some agreements (but also some disagreements) between the single-particle theory and the observed LLs.

Ref. [14] demonstrated that at zero flux, the second through fifth bands of the BM model, counting from CNP, form an elementary band representation and are forced to be connected by symmetry. Hence no gap is expected in the resistance data (Fig. 3b) between $\nu = 4$ and $\nu = 20$ at 0 flux, although there is a Dirac point at $\nu \sim 12$ leading to a low density of states (see Extended data F). This can be seen in Fig. 3b from the deep blue conducting regions near $B = 0$, which are punctuated by the less conductive (lighter blue) region near $\nu = 12$. The full-density Bloch bands at zero flux split into Hofstadter sub-bands upon applying a magnetic flux, giving rise to gaps at fractional fillings. The Chern number C of the gaps is given by the Streda formula: $n = pC \bmod q$, where N is the number of Hofstadter sub-bands that have been filled, as measured from CNP. At fractional filling ($n \neq 0 \bmod q$) C must be non-zero. When interactions, spin-orbit coupling, and the Zeeman effect are neglected, all bands are four-fold degenerate because of the spin and valley degeneracy.

From $\Phi = 0$ to $\Phi \sim 0.5\Phi_0$, there are three prominent features in the Wannier diagram (Fig. 3c) appearing at $v = 4$, 8, and 12. Near $v = 4$, we predict and observe positive slope LLs emerging from the band edge which can be attributed to the low-energy Rashba point in the passive bands of the zero flux BM model [28]. More interesting are the LLs which are connected to $v = 8$. Although there is no band edge at $v = 8$ in the BM model, the magnetic field breaks the C_{2x} and $C_{2z}T$ symmetries that enforce the second and third bands to be connected. Thus, we can understand the LLs pointing to $v = 8$ as indicating a nascent band edge which is revealed by flux in precise agreement with Fig. 3a., which demonstrates strong gaps originating from $v = 8$, with Chern numbers 4, 8, 12, 16. We expect increased R_{xx} in the regions where LLs of different slopes cross [28]. This is observed in Fig. 3b near $v = 10$ where the (8, 4) and (12, -4) LLs collide near $0.5\Phi_0$. Additionally, at $0.5\Phi_0$ flux, we observe very clean, highly conducting regions in Fig. 3b between the Chern gaps at $v = 8$, 10, 12, 14 and 16, corresponding to the metallic regions in the Hofstadter spectrum (see Extended data F), marked by light blue bars in Fig. 3a and 3c.

Some LLs in Fig. 3b and 3c with $v_L = 10$, 18, 22 (not divisible by 4) cannot be explained by the single-particle Hofstadter calculations and rely on interactions to break the spin-valley degeneracy. These LLs originate from the strongly interacting flat bands at zero flux and appear to remain competitive many-body states even at large flux and high fillings. There are also LLs with $v_L = 8$, 16 (divisible by 4) and 22 from $v = 4$ which do not appear in our single-particle calculations. Further work is necessary to characterize these states.

In summary, we report the first observation of interaction-driven correlated insulating phases at one flux quantum per moiré unit cell in MATBG. Our experimental observations largely agree with our single-particle Hofstadter calculations, which predict the emergence of a set of electronic flat bands at full flux with different symmetry and topology than the zero-field flat bands. These bands are unstable to the creation of correlated states by interactions [17].

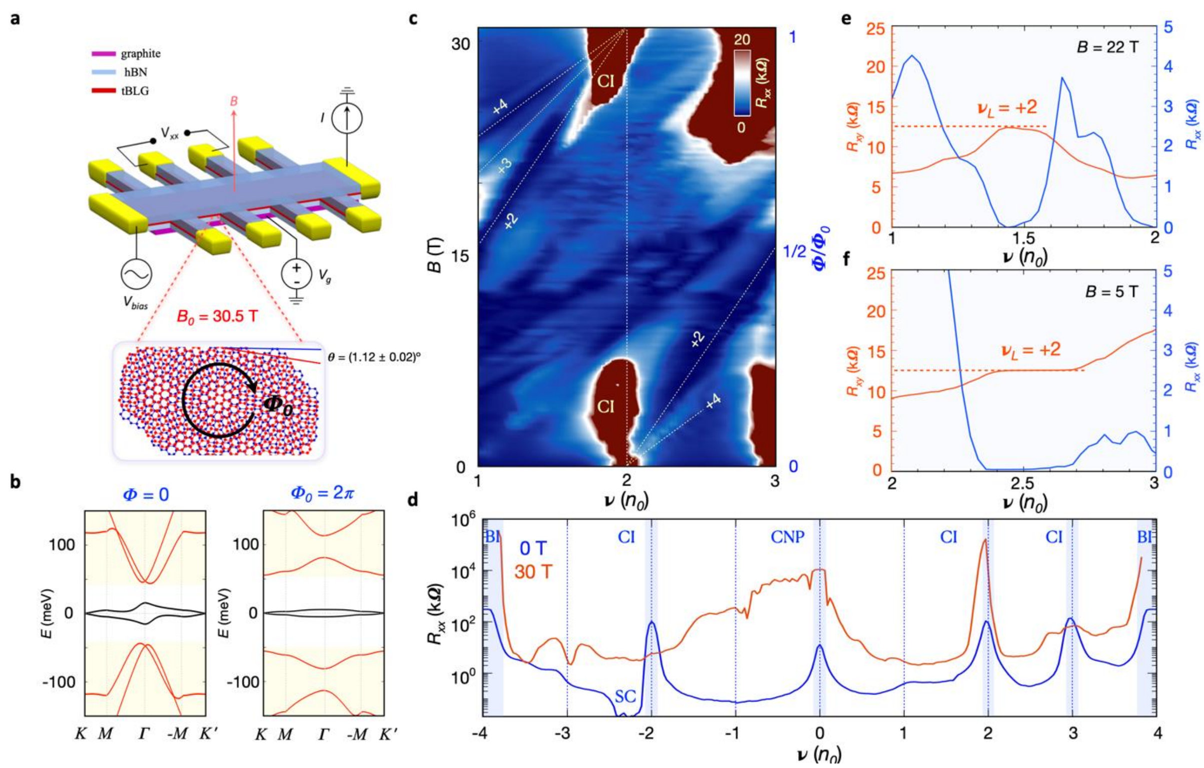

FIG. 1. **a)** Schematic of the MATBG device with the performed measurement scheme of the back-gated four-terminal R_{xx} and R_{xy} measurements. The twist angle of the device is $\theta = 1.12° \pm 0.02°$, which results in $B_0 = 30.5$ T. **b)** Band structure at zero and Φ_0 magnetic flux. **c)** Color plot of R_{xx} as a function of ν and B measured at $T = 40$ mK around ν = 2, showing the re-entrant CI and emergence of LLs. **d)** R_{xx} vs. ν over the entire range of the flat band at $B = 0$ T and $B = 30$ T, clearly showing the re-entrant CI at ν = +2 close to Φ_0. **e)** and **f)** R_{xx} and R_{xy} as a function of ν at B = 22 T and $B = 5$ T, respectively, showing full quantization of the $\nu_L = +2$ LL gap.

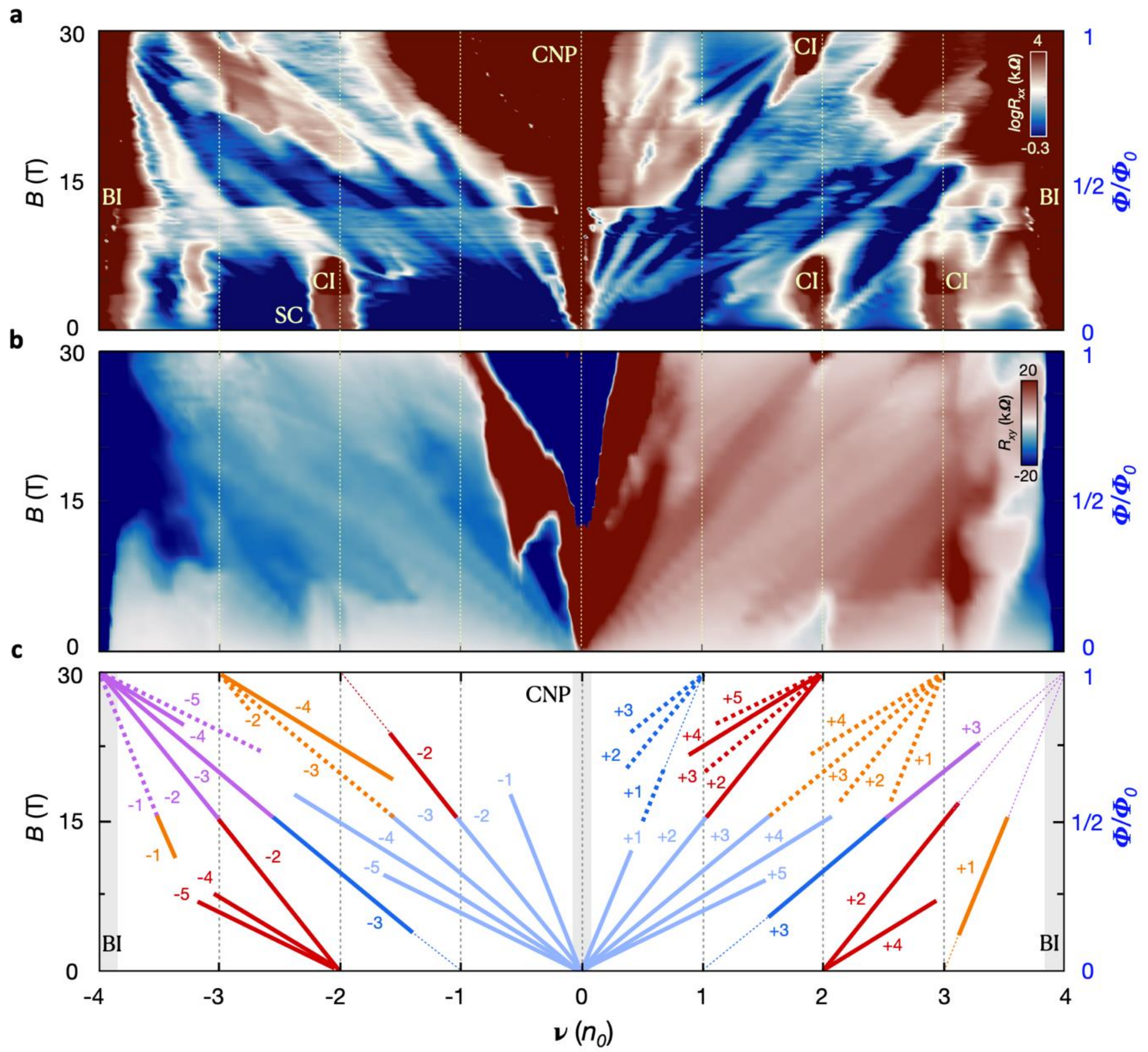

FIG. 2. a) and **b)** show respectively, the color plots of R_{xx} and R_{xy} as a function of B and ν, for the full magnetic phase space from B = 0 T to B = 31 T and ν from -4 to 4. **c)** Schematics of all the LLs emerging from different fillings of the band from both zero flux and Φ_0. Light blue lines from the CNP indicate the LLs with ν_L = ±1, ±2, ±3, ±4, ±5. Dark blue lines indicate the LLs from ν = ±1 at both zero flux and Φ_0. Dark red lines correspond to LLs from ν = ±2 at both zero flux and Φ_0. Orange lines indicate the LLs that emerge from ν = ±3 and purple lines correspond to LLs from ν = -4. Solid lines mark well-pronounced, quantized LLs, while dashed lines mark much weaker, non-quantized levels.

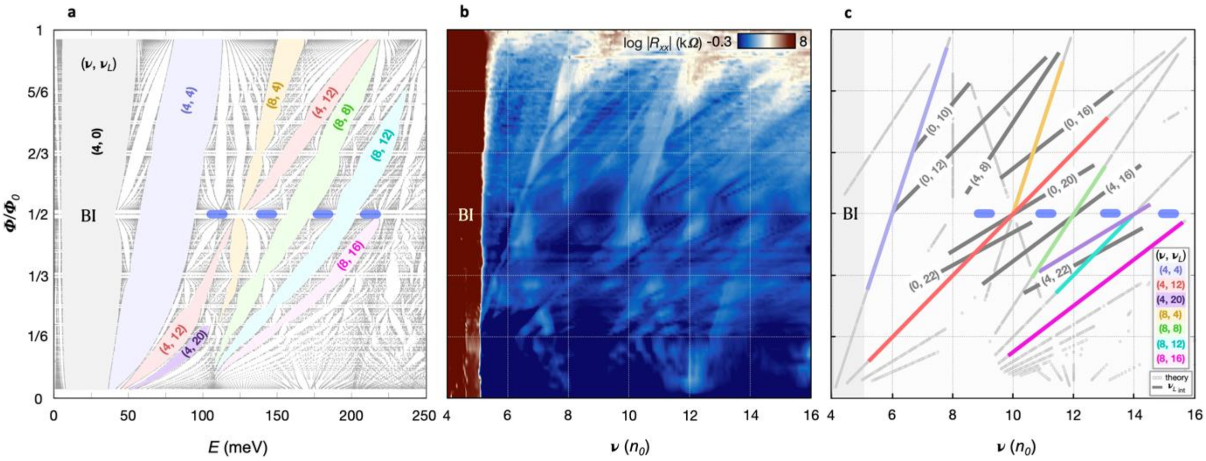

FIG. 3. a) Calculated Hofstadter spectrum of the BM model for positive energy as a function of the magnetic flux Φ through the moiré unit cell. Different solid grey (4, 0), blue (4, 4), red (4, 12), yellow (8, 4), green (8, 8), cyan (8, 12), magenta (8, 16) and purple (4, 20) regions correspond to the evolution of dominant LL gaps. They are marked with the band filling and filling factor of the LLs (ν, ν_L). **b)** Color plot of log(R_{xx}) as a function of B and ν for high range of carrier density up to $\nu = 16$. **c)** Schematics of (b) and the comparison with (a), where grey circles mark the theoretically predicted gaps from (a) and colored lines mark the strongest LLs in (b). The observed LLs from (b) are plotted with the same color code as the corresponding LLs in (a). Dark grey lines correspond to LLs, which are not predicted from the Hofstadter calculations. These levels are the result of strong interactions in the system (ν_{Lint}). The horizontal blue bars denote metallic regions in (a) matching the high conductance regions (dark blue regions) observed in (b).

Acknowledgements

D.K.E. acknowledges support from the Ministry of Economy and Competitiveness of Spain through the ‘Severo Ochoa ’programme for Centres of Excellence in R and D (SE5-0522), Fundacio Privada Cellex, Fundacio Privada Mir-Puig, the Generalitat de Catalunya through the CERCA programme, funding from the European Research Council (ERC) under the European Union’s Horizon 2020 research and innovation programme (grant agreement no. 852927). B. A. B.'s work was primarily supported by the DOE Grant No. DE-SC0016239, the Schmidt Fund for Innovative Research, Simons Investigator Grant No. 404513, and the Packard Foundation. Z.-D. S. was supported by ONR No. N00014-20-1-2303, NSF-MRSEC No. DMR-1420541, Gordon and Betty Moore Foundation through Grant GBMF8685 towards the Princeton theory program. B.A.B. also acknowledges support from the European Research Council (ERC) under the European Union's Horizon 2020 research and innovation program (Grant Agreement No. 101020833). I.D. acknowledges support from the INphINIT ‘laCaixa ' (ID 100010434) programme (LCF/BQ/DI19/11730030). K.W. and T.T. acknowledge support from the Elemental Strategy Initiative conducted by the MEXT, Japan (Grant Number JPMXP0112101001) and JSPS KAKENHI (Grant Numbers 19H05790, 20H00354 and 21H05233).

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