Hierarchy of Hofstadter states and replica quantum Hall ferromagnetism in graphene superlattices

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Self-similarity and fractals have fascinated researchers across various disciplines. In graphene placed on boron nitride and subjected to a magnetic field, self-similarity appears in the form of numerous replicas of the original Dirac spectrum, and their quantization gives rise to a fractal pattern of Landau levels, referred to as the Hofstadter butterfly. Here we employ capacitance spectroscopy to probe directly the density of states (DoS) and energy gaps in this spectrum. Without a magnetic field, replica spectra are seen as pronounced DoS minima surrounded by van Hove singularities. The Hofstadter butterfly shows up as recurring Landau fan diagrams in high fields. Electron-electron interactions add another twist to the self-similar behaviour. We observe suppression of quantum Hall ferromagnetism, a reverse Stoner transition at commensurable fluxes and additional ferromagnetism within replica spectra. The strength and variety of the interaction effects indicate a large playground to study many-body physics in fractal Dirac systems.

When graphene is placed on top of atomically flat hexagonal boron nitride (hBN) and their crystallographic axes are carefully aligned, the electron transport properties of graphene become strongly modified by a hexagonal periodic potential induced by the hBN substrate\textsuperscript{1,6}. Replicas of the main Dirac spectrum appear\textsuperscript{2,3} at the edges of superlattice Brillouin zones (SBZs) and, for the lowest SBZ, the second-generation Dirac cones can be reached using electric field doping\textsuperscript{4,6}. Because the superlattice period, $\lambda$, for aligned graphene-hBN structures is relatively large ($\sim$15 nm), magnetic fields $B \sim 10^5$ T are sufficient to provide a magnetic flux $\Phi$ of about one flux quantum $\phi_0$ per area $A = \sqrt{3}/2$ of the superlattice unit cell. The commensurability between $\lambda$ and the magnetic length $l_B$ gives rise to a fractal energy spectrum, the Hofstadter butterfly\textsuperscript{4,6,11-19}. An informative way to understand its structure is to consider the butterfly as a collection of Landau levels (LLs) that originate from numerous mini-replicas of the original spectrum, which appear at all rational flux values $\Phi = n \phi_0 (p/q)$, where $p$ and $q$ are integers\textsuperscript{11-13}. At these fluxes, the electronic spectrum can be described\textsuperscript{11-19} in terms of Zak’s minibands\textsuperscript{14} for an extended superlattice with a unit cell $q$ times larger than the original one. In graphene, Zak’s minibands are expected to be gapped cones (third-generation Dirac fermions)\textsuperscript{15}. Away from the rational flux values, these Dirac replicas experience Landau quantization in an effective field $B_{\text{eff}} = B - B_{\text{p/q}}$ where $B_{\text{p/q}} = q \phi_0 (p/q)/A$.

In this work, we have employed capacitance measurements to examine the electronic spectrum of graphene superlattices and its evolution into the Hofstadter butterfly. In zero $B$, pronounced minima in the electronic density of states (DoS) are observed not only for graphene’s neutral state but also at high electron and hole doping. The latter minima signify Dirac replicas near the edges of the first SBZ. The replicas occupy a spectral width of $\sim$50 meV, indicating strong superlattice modulation. The temperature ($T$) dependence of the DoS suggests that the second-generation Dirac cones are singly and triply degenerate for the valence and conduction bands of graphene, respectively. In quantizing $B$, in addition to the classic fan diagram for graphene, we observe many new cyclotron gaps falling out from finite values of $B$. They are attributed to the formation of high-field replica Dirac cones\textsuperscript{7} and their Landau quantization in $B_{\text{eff}}$. The local fans are particularly well developed near $\Phi = \phi_0$ and $\nu = 0, \pm 1, \pm 2$, where $\nu = n \phi_0 / B$ is the filling factor and $n$ the carrier density. The Hofstadter minigaps in this regime cannot be explained by orbital quantization alone. They do not follow the expected dependence on the LL index and are described by the Coulomb energy scale $E_C = e^2/\epsilon l_B^2$, where $l_B$ is the magnetic length in $B_{\text{eff}} = \pm |B - B_{\text{p/q}}|$ and $\epsilon$ the effective dielectric constant\textsuperscript{20-22}. We also observe that the SU(4) quantum Hall ferromagnetism (QHF), characteristic of non-aligned devices\textsuperscript{22-23}, experiences strong suppression at commensurable fluxes. The $\nu = \pm 1$ gaps disappear near $\Phi = \phi_0$, whereas the ferromagnetic states at $|\nu| = 3, 4$ and 5 exhibit a re-entrant transition at $\Phi = \phi_0/2$.

**DoS for second-generation Dirac fermions**

Figure 1a shows our capacitor devices. Graphene is placed on top of hBN (50–100 nm thick) and encapsulated with a second hBN.
Figure 1 | Capacitance spectroscopy of graphene superlattices. a, Typical C(V_b) in zero magnetic field. Positive and negative V_b correspond to electron and hole doping, respectively. The range of applied V_b is limited by the dielectric breakdown of hBN (≤0.5 eV nm^-1). For the shown capacitor, S = 308μm^2 and d = 35 nm. Right insets: Schematics of our devices and their optical image. The two square regions are Au gates, and the top and bottom leads are electrical contacts to graphene. Scale bar, 20 μm. Left inset: One of the theoretically proposed scenarios for the low-E band structure of graphene on hBN (refs 11,12). The first, second and third SBZ are shown in red, green and blue, respectively. b, Density of states (DoS) for the device in a. The inset shows the T dependence of the DoS in the three minima (blue squares, main Dirac point (DP); solid red circles, hole-side DP; open red circles, electron-side DP). Solid curves: theoretical fits. Note that smearing of the DoS by scattering and inhomogeneity is less than 50 K.

crystal of thickness d. A gold electrode is then evaporated on top. The whole structure is fabricated on a quartz substrate to minimize parasitic capacitances. The devices are similar to those studied previously, but a critical step is added: crystallographic alignment of graphene and one of the encapsulating hBN crystals with a precision of ~1° using procedures described in ref. 4. Seven capacitors have been studied, with areas S ranging from 50 to 350 μm^2 and d ranging from ~10 to 40 nm. Depending on accuracy of our alignment, the densities n at which the first SBZ becomes fully capped are found between ~3 and 6 × 10^11 cm^-2. All the capacitors studied exhibit slight residual doping (Fig. 1a), and their charge carrier mobilities vary from ~50,000 to 120,000 cm^2 V^-1 s^-1, as found using Hall bar devices fabricated in parallel with the capacitors. The differential capacitance C was measured using an on-chip bridge made following the method described in ref. 6. Note that no Hořová states were observed in previous capacitance measurements of graphene superlattices.

The typical behaviour of C in zero B as a function of bias V_b, applied between graphene and the Au electrodes, is shown in Fig. 1a. A sharp minimum near zero V_b corresponds to the main neutrality point where the DoS tends to zero and remains finite only as a result of charge inhomogeneity. There are additional minima at large electric-field doping (Fig. 1a). Following the earlier analysis of the features can be attributed to second-generation Dirac cones (inset in Fig. 1a). It is possible to translate C directly into the DoS or, equivalently, the quantum capacitance C_Q = C d x DoS, where DoS is the density of states and e the electron charge. To this end, we write 24

\[ C = (1/C_{G} + 1/C_{p})^{-1} + C_{c}, \]

where \( C_{G} \) and \( C_{p} \) are the geometric and parasitic (parallel) capacitances, respectively. \( C_{c} \) is defined by the known values of d and S and can be measured independently using the periodicity of the magnetocapacitance oscillations24. This leaves \( C_{c} \) as the only fitting parameter to determine \( C_{G} \). In our devices, \( C_{c} \approx \sim 1 \text{fF (}< 1\% \text{ of } C) \) and, in most cases, no fitting is required. To present the DoS as a function of the Fermi energy E rather than V_b, we first integrate C(V_b) curves over V_b, which yields the induced n, and then subtract the electrostatic voltage drop from the applied bias24: E = eV_b - e n / C_G (again, no fitting parameters). Examples of this conversion of C(V_b) into DoS(E) are shown in Fig. 1b. In this presentation, the spectral changes induced by the superlattice potential become clear and more pronounced. Instead of the standard linear behaviour (DoS \( \propto |E| \)) seen for non-aligned graphene-on-hBN capacitors, deep minima in the DoS appear for \( |E| > 0.15 \text{ eV} \) in both the valence and conduction bands of graphene. The minima are surrounded by equally pronounced maxima. The behaviour is a manifestation of the Dirac replicas formed at the edges of the first SBZ and terminated by van Hove singularities (left inset of Fig. 1a). This agrees with the earlier results obtained by scanning tunnelling spectroscopy, where qualitatively similar but more smeared DoS curves were observed. The spectral width occupied by the second-generation Dirac fermions (distance between van Hove singularities) is ~25 and ~75 meV for positive and negative E, respectively. The non-monotonic behaviour at \( |E| > 0.2 \text{ eV} \) is reproducible for different devices and indicates contributions coming from further SBZ.

Taking into account graphene's spin and valley degeneracy, we write DoS = 8πg_s |E| / \( h^2 v_F^2 \), where \( h \) is the Planck constant and \( g_s \) the additional degeneracy of secondary Dirac fermions (inset of Fig. 1a). The observed linear behaviour at \( |E| < 0.1 \text{ eV} \) in Fig. 1b yields \( v_F = 0.98 \pm 0.04 \times 10^8 \text{ m s}^{-1} \), in agreement with literature values. The slopes DoS \( \propto |E| \) extrapolate to zero DoS with no indication of a sizeable gap (>5 meV) opened by the hBN potential. Further information about secondary Dirac fermions can be obtained by analysing the temperature dependences of the DoS (Fig. 1b). At a Dirac point (DP), the DoS is expected to increase linearly with T as DoS(T) = 8πln(4g_s)T / \( h^2 v_F^2 \). For the main spectrum, \( g_s/v_F^2 \) is known and no fitting parameter is needed to describe the observed behaviour at high T (inset of Fig. 1b). The saturation at low T is due to charge inhomogeneity, which we model by the Gaussian distribution of n with a standard deviation \( \delta n \). The blue curve in the inset of Fig. 1b is for \( \delta n = 1.3 \times 10^6 \text{ cm}^{-2} \), in agreement with the smearing of DoS as a function of E at low T. For hole- and electron-side replicas, their DoS at high T increases by approximately 5 and 15 times, respectively, quicker than that at the main minimum. By taking into account that V_b is expected to be \( \sim v_F^2 / 2 \) and assuming that \( \delta n \) does not change significantly with V_b, the data in the inset of Fig. 1b yield \( g_s = 1 \) and \( v_F \approx 0.45 \times 10^8 \text{ m s}^{-1} \) for the hole-side Dirac cones and \( g_s = 3 \) and \( v_F \approx 0.4 \times 10^8 \text{ m s}^{-1} \) for the electron-side Dirac cones (red curves). These values are also consistent with the observed DoS(E) curves. The values of \( g_s \) found imply that there are one and three Dirac replicas for negative and positive E, respectively, in agreement with the spectral reconstruction scenario shown in the inset of Fig. 1a.
Capacitance spectroscopy of the Hofstadter butterfly

Figure 2 shows examples of $C(V_b)$ in the regime of quantized $B$. Landau quantization results in numerous sharp minima, which develop in $B > 1\, \text{T}$, so that at $3\, \text{T}$ one can see all the cyclotron gaps for $v$ up to 38 (Fig. 2a). Many-body gaps at $v = 0$ and ±1 (owing to the lifted spin-valley degeneracy) open up at $\sim 1.5$ and $4\, \text{T}$, respectively. In low $B$, we can employ the same approach as described above to convert $C(V_b)$ into DoS ($E$). Examples are given in Fig. 2d and Supplementary Fig. 1. One can see sharp peaks in the DoS which correspond to metallic LLs (incompressible states with $C \approx C_0$) separated by wide regions of a low DoS (cyclotron gaps). The conversion procedure cannot be applied automatically to strongly quantizing $B$ because in the quantum Hall regime the bulk becomes increasingly isolated from electrical contacts, and central areas no longer contribute to the measured signal. This leads to excessively deep minima in $C$ (effectively, smaller $S$) and large systematic errors in determining $E$ (Supplementary Section 1). We can increase $T$ to suppress the insulating state, but because of different gap sizes this has to be done individually for each gap and each range of $B$ (Supplementary Fig. 1). Therefore, our high-$B$ data are mostly presented in the original format, as $C(V_b)$.

In addition to the cyclotron gaps described above, we find numerous magnetocapacitance minima that are not observed in similar but non-aligned graphene-on-hBN devices$^{13}$. The minima become very pronounced in high $B$ (Figs 2b–c), and their evolution leads to the complex pattern seen in Fig. 3a. For clarity, the pattern is reproduced schematically in Fig. 3b. The observed fan diagram is consistent with those reported previously$^{14,15}$ but, rather unexpectedly, our capacitance measurements reveal more Hofstadter states than could be seen in the transport experiments.

The colours in Fig. 3b indicate the four types of gap. First, there are the expected single-particle gaps originating from the main DP (black lines). Second, there are many-body gaps due to QHFM, indicated by the blue lines. Third, there are minima in the DoS which fan out from hole- and electron-side DPs (green). Intuitively, one could attribute these to LLs for second-generation Dirac fermions. However, this interpretation is wrong because it is easily estimated that the cyclotron energy already becomes comparable to the spectral width of the Dirac replicas for $B < 1\, \text{T}$. Accordingly, all the LLs should bunch at van Hove singularities (Fig. 1b) before even being resolved in our measurements. Comparison with theory$^{4,12}$ suggests that the green gaps represent Hofstadter minibands in the regime of strong superlattice modulation, where any distinction between LLs originating from the main and superlattice spectra is lost. Fourth, our fan diagrams show minima that cannot be traced back to either the main or secondary DPs (red). We attribute them to LLs originating from replica Dirac spectra that are quantized in $B_{\phi} = \pm |q(B_{\phi} - B)|$ with $q = 1, 2, \ldots$. This is seen most clearly around $\Phi = \phi_0$, where minigaps spray up and down from $v = 0, \pm 1, \pm 2$ and −6 (Fig. 3). This is a case of relatively weak modulation such that individual LLs split into superlattice minibands without intermixing$^{16-18}$. As discussed above, the superlattice band structure at exactly $\Phi = \phi_0$, consists of replica Dirac cones$^{12}$ and, for both positive and negative $\phi_0$, these cones experience quantization that leads to local fan diagrams. Note that the red and green gaps in Fig. 3b have essentially the same (that is, superlattice) origin and we distinguished between them above only to facilitate the discussion.

Interaction effects in the Hofstadter spectrum

The simultaneous occurrence of QHFM and Hofstadter states suggests a possibility of their interplay. Several observations show that this is indeed the case. Most striking among them is the suppression of QHFM at commensurable fluxes $\Phi/\phi_0 = 1$ and 2/1 (Fig. 4). One can notice this effect already in Fig. 3, where the QHFM gaps at $v = 3, \pm 4, \pm 5$ disappear around $\Phi = \phi_0/2$ (also, see Supplementary Fig. 4). The suppression of QHFM is further elucidated in Fig. 4bc. After the onset of QHFM in a field of a few $\text{T}$, the main many-body gaps grow gradually with increasing $B$ but then shrink approaching $\Phi = \phi_0$. The QHFM gaps at $v = \pm 1$ vanish around $\Phi = \phi_0$, and there are weaker minima near $\Phi = \phi_0/2$ (clearly developed for $v = -1$; see Fig. 4c). This behaviour has not been observed in graphene-on-hBN without alignment$^{24}$ (see Supplementary Information).

We attribute the suppression of QHFM to a reverse Stoner transition. According to numerical modelling over a broad range of moiré parameters$^{41,42}$, the zero LL remains relatively narrow for $B < 0.7B_{11}$ but then broadens exponentially into a sizeable band of width $D$ (Supplementary Fig. 5). As the spin-valley ferromagnetism specifically requires narrow LLs, the superlattice broadening of a LL should result in a tendency of the system to return to the normal metal state or, at least, reduce the associated QHFM gap by $D$. In moderate $B$, the QHFM gap at $v = 0$ is accurately described by $E_C = e^2/\ell_b \propto \sqrt{B}$ (inset of Fig. 4c). By extrapolating this behaviour to high $B$ (black curve in Fig. 4c), we can estimate $D$ of the zero LL from the relative drop in the measured gap, $\Delta = E_C - D$. This yields $D \approx 5\, \text{meV}$ at $\Phi \approx \phi_0$. The complete suppression of the QHFM at...
\(\nu = \pm 1\) is attributed to the low energy costs of creating skyrmion-like spin/valley textures, which in combination with sizeable \(D\) close the gaps (Supplementary Section 4).

Figure 4 provides a closer look at incompressible superlattice states \((\nu, \nu_l)\) near \(\Phi = \Phi_0\). One may naively attribute them to cyclotron gaps for third-generation Dirac fermions in fields \(B_{\text{eff}} = B - B_{\text{LL}}\). However, states \((\pm 2, \nu_l)\) fanning out from \(\nu = \pm 2\) contain pronounced gaps at odd \(\nu_l\) (Fig. 4a). Because the \(\nu = \pm 2\) quantum Hall states are neither spin nor valley polarized, one cannot explain these odd-integer gaps simply by superlattice effects, without invoking spontaneous lifting of the degeneracies. Furthermore, the inset in Fig. 4c shows one of the main odd-integer minigaps \((-2,1)\) as a function of \(B_{\text{eff}}\) and compares it with the QHFM gap at \(\nu = 0\) plotted as a function of \(B\) (the latter's behaviour agrees with the earlier report\(^{36}\)). Both gaps \((\Delta)\) exhibit absolute values close to \(E_C = e^2/\ell_B \propto \sqrt{B}\), where \(\ell_B = (h/2\pi e B)\) and \(B\) is given by either \(B_{\text{eff}}\) or \(B_{\text{LL}}\) (Fig. 4a). This agreement indicates the two gaps have a common origin. The above reasoning applies also to the fan diagram \((0, \nu_l)\), which contains odd \(\nu_l\). The odd-integer minigaps \((0, \pm 1)\) exhibit the same dependence \(\Delta \propto \sqrt{B_{\text{eff}}}\) but smaller absolute values. Our analysis of the effect of electron-electron interactions on LLs of third-generation Dirac fermions is given in Section 4 of the Supplementary Information and provides a qualitative understanding of the entire hierarchy of observed minigaps. For example, the replica quantum Hall ferromagnetism allows us not only to understand the presence of the large gaps at \((-2,1)\) and \((-2,2)\) but also the virtual absence of similarly indexed states at \((-2,-1)\) and \((-2,-2)\) (see Fig. 4a).

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**Figure 3 | Hofstadter butterfly in graphene superlattices.** a | Fan diagram \(C(V_b, B)\). Gaps appear as dark stripes. Scale: navy-to-white, 0.9–1.02 \(C_G\), black, \(C < 0.9C_G\). Such zooming does not distinguish between deep and shallow gaps, which both appear dark. The data are for the device in Fig. 2. Our other superlattice capacitors show similar behaviour, but the weakest minima may disappear owing to charge inhomogeneity (Supplementary Section 2). The measurements were done in a superconducting magnet (up to 18 T) and in an electromagnet at the High Magnetic Field Laboratory at Grenoble (up to 29 T). The presented diagram is a combination of the two sets. Data within the red dashed rectangle \((|\nu| < 2)\) were accumulated over several days to improve the resolution. b | Density of states (DoS) minima from a shown schematically. The grey lines are a Wannier grid, showing the allowed gaps, including those with lifted spin and valley degeneracies. \(4 \times n_g\) is the carrier density at which the first superlattice Brillouin zone is filled. \(\Phi = B \times A\) for the right axis is calculated using the known value of \(n_g = 1/A\), which yields \(A\) directly\(^{4-6}\).

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**Figure 4 | Interactions in Hofstadter minibands.** a | Zoom into the high-\(B\) region of Fig. 3a. The diameters of the yellow circles indicate the relative sizes for superlattice minigaps at 29 T. They are marked as \((\nu, \nu_l)\), where the filling factor \(\nu\) indicates the association with the \(\nu\)-th gap in the main spectrum and \(\nu_l\) is the local filling factor. The latter is determined from the slopes of \(B(n)\) in the fan diagram in Fig. 3b, which yield a value of \(\nu_l - \nu\) (Supplementary Information). b | Cross-sections from a to illustrate the closure of the \(\nu = \pm 1\) gaps around 25 T. c | Evolution of the main spin-valley gaps \((\Delta)\) with \(B\). The gaps are obtained by integration of the \(C(V_b)\) curves. Inset: Largest minigap \((-2,1)\) as a function of \(B_{\text{eff}}\) (red symbols) and the \(\nu = 0\) gap as a function of \(B\) (blue). The gaps are found from fitting the temperature dependences of the capacitance minima at fixed \(B\) using the Lifshitz–Kosevich formula\(^{44}\) (the bars show the standard error for the fitting). Both gaps trail the Coulomb energy scale \(E_C\) with \(e = 8\) for encapsulated graphene\(^{44}\) (solid curve). \(T = 2\ K\) for all the plots.
Outlook
Capacitance spectroscopy is shown to be a powerful and, at the same time, relatively easily accessible tool for investigating complex graphene systems. The possibility of converting capacitance data directly into the DoS makes the approach even more appealing. The variety of many-body phenomena observed in graphene superlattices is impressive and merits further studies. We expect they will reveal new skyrmion physics within Hofstadter states and, as the quality of graphene superlattices improves, replicas of fractional quantum Hall effect states should also become observable.

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Author contributions
J.S.T., A.V.K., Y.C., R.J. and F.W. designed and fabricated the devices. A.M. and G.L.Y. carried out the measurements. B.A. and M.P. helped with high-field experiments. X.C. and V.I.F. provided theoretical support. K.W. and T.T. provided HN crystals. R.X.G. devised the fabrication technology for graphene capacitors. A.M. developed the on-chip capacitance bridge. A.M., V.I.F. and A.K.G. analysed the results. A.K.G. together with J.S.T., A.V.K., Y.C., R.J. and F.W. designed and fabricated the devices. A.M. and G.L.Y. contributed to discussions.

Additional information
Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to V.I.F., A.K.G. or A.M.

Competing financial interests
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