Cloning of Dirac fermions in graphene superlattices

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Superlattices have attracted great interest because their use may make it possible to modify the spectra of two-dimensional electron systems and, ultimately, create materials with tailored electronic properties1–4. In previous studies (see, for example, refs 1–8), it proved difficult to realize superlattices with short periodicities and weak disorder, and most of their observed features could be explained in terms of cyclotron orbits commensurate with the superlattice4–6. Evidence for the formation of superlattice minibands (forming a fractal spectrum known as Hofstadter’s butterfly7) has been limited to the observation of new low-field oscillations8 and an internal structure within Landau levels9–11. Here we report transport properties of graphene placed on a boron nitride substrate and accurately aligned along its crystallographic directions. The substrate’s moiré potential8–10 acts as a superlattice and leads to profound changes in the graphene’s electronic spectrum. Second-generation Dirac points12–22 appear as pronounced peaks in resistivity, accompanied by reversal of the Hall effect. The latter indicates that at the main neutrality point, whereas that on the electron side is 10 times weaker. The reversal in sign of ρxy (Fig. 1b) cannot be explained by additional scattering and proves that hole-like and electron-like carriers appear in the conduction and, respectively, valence bands of graphene. We attribute the extra neutrality points to the superlattice potential induced by the hBN, which results in minibands featuring isolated secondary Dirac points (Fig. 1a, inset). This interpretation agrees with theory12–22 and the tunnelling features reported in ref. 12, including the fact that those were stronger in the valence band.

Near the main neutrality point, the aligned devices have transport characteristics typical for graphene on hBN27–28. The conductivity σ(n) = 1/ρxx varies linearly with n and can therefore be described in terms of constant mobility, μ. For the reported devices, we find that μ = (20–80) cm2 V−1 s−1 for |n| > 1011 cm−2. Around the secondary neutrality point, σ depends linearly on n – nS. At low temperatures T, the secondary peak on the hole side is stronger than that at the main neutrality point, whereas near the electron-side secondary neutrality point we find even higher values, μ = (30–100) cm2 V−1 s−1. However, at the main and secondary neutrality points the T dependences of both μ and the minimum conductivities are different. This is discussed in Supplementary Information, section 1, and here we note only that the observed functions σ(T) do not support the idea of major energy gaps being induced by the superlattice at the cloned secondary Dirac points19–22 (Fig. 1a, inset). Furthermore, following the approach described in ref. 29, we analysed the thermal broadening of the peaks in ρxx (Supplementary Information, section 2). The analysis proves that the spectrum at the secondary neutrality points is linear, that is, Dirac-like, in agreement with theory12–22. Figure 2 shows the evolution of ρxx(n) with increasing perpendicular magnetic field, B. Near the main Dirac point, we observe the standard26 quantum Hall effect (QHE) for graphene, with plateaux in ρxy and zeros in ρxx at filling factors ν = nφ0/ 2π = ±2, ±6, ±10, . . . where φ0 is the flux quantum. Fan diagrams around the secondary Dirac points are different (Fig. 2). The resistance peak of the hSNP first broadens with increasing B and then splits into two maxima. The maxima correspond...
to the superlattice filling factors $v_S = \pm 2$, where the carrier density is counted from the hSNP. In the middle of each maximum, there is a deep minimum (narrow white stripes in Fig. 2a). The minima in $\rho_{xx}$ are accompanied by positive and negative extrema in $\rho_{xy}$ (Fig. 2c, d). This shows that electron-like cyclotron trajectories in graphene’s valence band persist when $B$ is quantizing (that is, when it quantizes the spectrum). With decreasing $T$, $\rho_{xx}$ inside the narrow minima tends to zero and the corresponding extrema in $\rho_{xy}$ become increasingly more pronounced, which is behaviour characteristic of the development of Shubnikov–de Haas oscillations into QHE states (Fig. 2c, d). The $T$ dependence yields a QHE gap of $\sim 20$ meV (Supplementary Fig. 6).

Unlike cyclotron gaps, this one is practically independent of $B$, as is seen also from the fact that the white stripes in Fig. 2a do not widen. With increasing $T$, the QHE states at $v_S = \pm 2$ gradually disappear below $50$ K but the maxima in $\rho_{xx}$ persist up to $150$ K.

Another notable feature of the observed fan diagrams are the multiple peaks in $\rho_{xx}$ accompanied by zeros or deep minima in $\rho_{xy}$. This is seen most clearly for devices where doping sufficiently higher than $n_S$ can be achieved (Fig. 3). Furthermore, in all our devices near the hSNP, $\rho_{xy}$ repeatedly changes its sign with increasing $\nu$, indicating recurring appearance and disappearance of electron-like orbits within graphene’s valence band (Fig. 3b, e and Supplementary Figs 4 and 5). This means that, for a given $n$, the magnetic field alone can repeatedly generate new neutrality points. Such ‘third-generation’ neutrality points occur periodically as $n$ and $1/B$ vary, and form distinct groups characterized by particular values of $1/B$ (Fig. 3 and Supplementary Information, section 3). Their periodicity in $1/B$ is accurately described by unit fractions, $\phi_0/q$, of the magnetic flux, $\Phi = B S_{\perp}$, per superlattice unit cell area, $S_{\perp}$, where $q$ is integer. In the conduction band, the fan diagrams also exhibit Landau levels extending from the secondary Dirac point, and numerous third-generation neutrality points with the same $1/B$ periodicity are visible in Figs 2b and 3a, b. These features are weaker than those in the valence band. For example, the resistivity peak at the electron-side neutrality point is no longer there for $B \approx 1$ T (Fig. 2b) and we did not observe the secondary QHE in the conduction band.

The observed superlattice behaviour suggests that complex spectral changes are induced by quantizing $B$. Theoretically, the problem is...
somewhat similar to that originally discussed by Zak and Hofstadter and later considered for two-dimensional electron systems in semiconductor superlattices and for Dirac fermions in twisted bilayers. The most general, but not proven, prediction is that superlattice spectra should be ‘self-similar’; that is, they should consist of multiple clones of an original spectrum, which appear at values of $B$ such that $\Phi = \phi \Phi / q$, where both $p$ and $q$ are integer. Our case of graphene on hBN is analysed in Supplementary Information, and the main theoretical results are summarized in Fig. 3c, d.

Figure 3c shows that the superlattice potential results in additional structure within each Landau level, which effectively broadens them with increasing $B$. The structured Landau levels extending from the main and secondary Dirac points strongly mix at high doping, $|E| \approx E_0 = \hbar v_F(\pi n_0)^{1/2}$. The resulting pattern is different from that in semiconductor superlattices with a parabolic spectrum and weak modulation. In the latter case, the fractal structure within each Landau level can be described by the original Hofstadter butterfly, which appears periodically as a function of $\phi \Phi / \Phi$. In our case, in which there is a Dirac-like spectrum and strong modulation, the fractal pattern depends on the Landau level index, $N$, and $B$ (Supplementary Information, section 5).

The calculated spectrum allows us to understand many features observed experimentally. Indeed, Fig. 3c shows a self-similarity such that magnetic states tend to entwine at $\Phi = \phi \Phi / q$, forming the fractal structure of the pattern. The strongest entwining occurs for unit fractions (that is, $p = 1$), and this results in an overall periodicity in $1/B$ with a period of $S_0/\phi_0$, in agreement with the experiment (Fig. 3a, b). The periodicity can be traced to the fact that for $\Phi = \phi \Phi / q$ the system can be considered a new superlattice that has a unit cell $q$ times larger than the original and that is placed in zero effective magnetic field. An example of the resulting magno-electronic (Zak) bands is given in Supplementary Fig. 8.

We find that in our case Zak bands feature slightly gapped Dirac spectra. This finding is illustrated in Fig. 3d, which, as an example, magnifies a part of Fig. 3c near the hSNP and $\Phi = \phi \Phi / 2$. Using the Zak spectrum calculated for $\Phi = \phi \Phi / 2$, we can obtain its Landau quantization in small reduced fields, $\delta B = B - B_2$, where $B_2 = \phi_0/2S_0$ is the zero effective field acting on the cloned Dirac spectrum. The shape of the resulting Landau levels is given by $\pm \sqrt{N|\delta B| + A^2}$, where $A$ is the gap in the local Dirac-like spectrum (Fig. 3d, magenta curves). Outside the empty horizontal region inaccessible in our numerical calculations (Supplementary Information, section 5), the Landau quantization of the Zak spectrum in $\delta B$ yields practically the same electronic states as shown by the calculated points (Fig. 3d, black dots). Similar local Dirac spectra are found in other parts of the moiré minibands for the case of four holes per moiré supercell (that is, at the hSNP) and found that $\Phi = \phi \Phi / q$ the Fermi energy lies inside the
corresponding Zak minibands, whereas for \( \Phi = q(\pi + 1/2) \) it lies inside gaps. This explains the experimentally observed oscillations in \( \sigma_{xx}(B) \).

Another experimental feature revealing Zak minibands and the hierarchy of superlattice gaps are the prominent QHE gaps at \( \nu = \pm 2 \) near the hSNP (Fig. 2). For small \( B \), they can be considered a result of Landau quantization for secondary Dirac fermions, and their zero Landau levels become separate from the rest of the spectrum (Fig. 3c). For higher \( B \), the resulting gaps saturate, being limited in size by the presence of van Hove singularities at the edges of the SBZ (Fig. 1a). As Zak minibands become increasingly more pronounced the secondary Landau levels intertwine with main Landau levels and, at high doping, become indistinguishable from them (Fig. 3c). Therefore, the complex pattern of Landau levels in Fig. 3 at high doping can no longer be interpreted in terms of Landau quantization at high doping, become indistinguishable from them (Fig. 3c).

The pattern becomes a ‘Hofstadter–Landau’ butterfly, specific to our strong-modulation regime and the linear spectrum. The largest fractal gaps near the hole-side Dirac point in Fig. 3c are in agreement with the \( \nu = \pm 2 \) QHE states observed experimentally, which have activation energies almost independent of \( B \). This behaviour is different from the case of weak modulation in semiconductor superlattices\(^6\), where Landau levels become structured but do not intertwine. However, increasing \( B \) such that \( \Phi > \phi_0 \) can drive graphene superlattices into the regime of weak modulation (Supplementary Fig. 9). This regime is outside the range of \( B \) values available in our experiment. In addition to the large fractal gaps, our experimental data also reveal reproducible small-scale structure that cannot be traced back to either main or secondary neutrality points (see, for example, Fig. 3a, b near the hSNP at high \( B \) and Supplementary Fig. 5a). We attribute these fine features to further fractalization of the superlattice spectrum such that isolated Landau levels for the third-generation Dirac clones start being resolved (Fig. 3d). This is similar to the intra-Landau-level features reported in semiconductor superlattices\(^6\) and warrants further investigation.

Graphene superlattices can be reliably fabricated for various types of transport measurement. This opens new lines of enquiry; in particular, the fractal quantization leads to such rich behaviour that its full understanding will require much further work, both theoretical and experimental. The demonstrated possibility of creating gaps at specifically chosen energies by controllably rotating graphene or other two-dimensional crystals within van der Waals heterostructures\(^2\) can be used to design novel electronic and optoelectronic devices.

**METHODS SUMMARY**

Our devices were multiterminal Hall bars fabricated following the procedure described in ref. 27. In brief, monolayer graphene was deposited on top of a relatively thick (\( > 30 \) nm) hBN crystal\(^2\) and then covered with another hBN crystal. The encapsulation protects the graphene from the environment and allows high-quality measurements at room temperature. To align the crystal lattices, we used an optical microscope that served as a back gate. To align the crystal lattices, we used an optical microscope that served as a back gate. To align the crystal lattices, we used an optical microscope that served as a back gate.

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**Supplementary Information** is available in the online version of the paper.

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