

## CONDENSED MATTER PHYSICS

## Minibands in twisted bilayer graphene probed by magnetic focusing

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Magnetic fields force ballistic electrons injected from a narrow contact to move along skipping orbits and form caustics. This leads to pronounced resistance peaks at nearby voltage probes as electrons are effectively focused inside them, a phenomenon known as magnetic focusing. This can be used not only for the demonstration of ballistic transport but also to study the electronic structure of metals. Here, we use magnetic focusing to probe narrowbands in graphene bilayers twisted at  $\sim 2^\circ$ . Their minibands are found to support long-range ballistic transport limited at low temperatures by intrinsic electron-electron scattering. A voltage bias between the layers causes strong minivalley splitting and allows selective focusing for different minivalleys, which is of interest for using this degree of freedom in frequently discussed valleytronics.

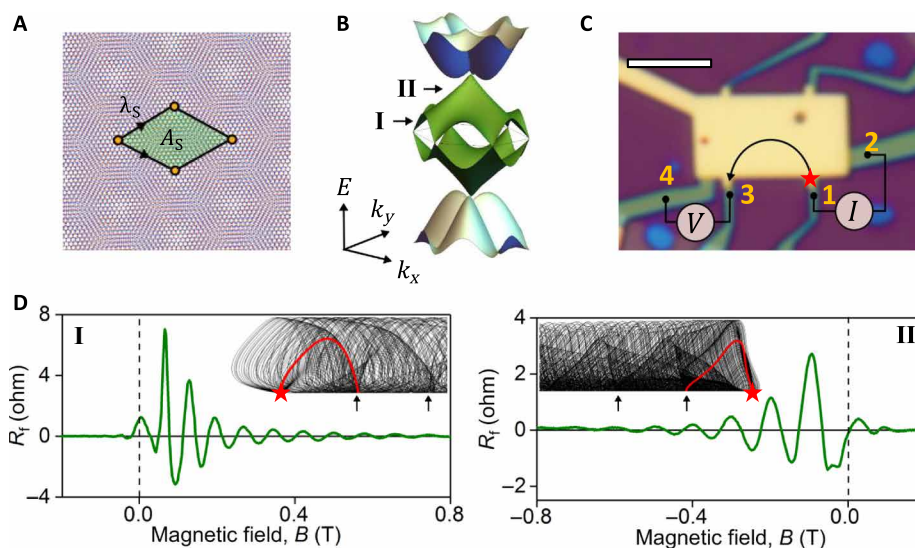
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## INTRODUCTION

Crystallographic alignment of atomically thin crystals stacked together in a van der Waals heterostructure is a powerful tool that enables fine-tuning of their electronic spectra. For crystals with similar honeycomb lattices, the spectra are modified by the presence of a long-range interference (moiré) pattern with a period  $\lambda_S$  dependent on the twist

angle  $\theta$  between the layers (see Fig. 1A) (1–18). The additional spatial periodicity reduces the size of the Brillouin zone and introduces secondary Dirac points, as illustrated in Fig. 1B. So far, the most pronounced twist-engineered changes in the electronic properties of two-dimensional (2D) crystals have been achieved in twisted bilayer graphene (TBG), where the twist at discrete “magic” angles results in narrowbands, periodically modulated interlayer hybridization, and strong enhancement of electron correlations, leading to superconductivity and Mott insulator transitions (6–8). At larger  $\theta$ , the TBG spectrum corresponds to a metal with several minibands at each K and K' valley in the Brillouin zone (Fig. 1B). Electronic properties of such a metal are expected to be quite different from

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**Fig. 1. Moiré minibands and TMF measurements.** (A) Schematics of the moiré superlattice induced by the twist of graphene layers. Here, two graphene sheets are rotated by an angle  $\theta$  relative to each other, which creates an additional spatial periodicity  $\lambda_S = a/[2 \sin(\theta/2)]$  ( $a$  is graphene's lattice constant) with the unit cell area of  $A_S = \sqrt{3}/2 \lambda_S^2$ . (B) Band structure of TBG graphene in the K valley of the Brillouin zone calculated for the twist angle  $\theta = 1.87^\circ$ , as discussed in section S3. (C) Optical image of TBG device D1 with  $\theta = 1.87^\circ$ . Scale bar, 4  $\mu\text{m}$ . (D) Two examples of TMF signals measured in device D2 ( $D = 0 \text{ V nm}^{-1}$ ) at 5 K for the carrier density  $3.7 \times 10^{12} \text{ cm}^{-2}$  (left) and  $9.3 \times 10^{12} \text{ cm}^{-2}$  (right) at a distance of 4.9  $\mu\text{m}$  from the injector. The latter is close to the main and secondary neutrality points, respectively, as illustrated in (B). The insets are examples of focusing caustics near the main (left) and secondary (right) neutrality points (see more examples in fig. S4). Arrows highlight the focal points for caustics, red star marks the current injection point, and red lines show typical trajectories that extend from the injector to the first focal point.

the behavior of Dirac electrons in monolayer or bilayer (aligned to Bernal stacking) graphene but so far remain largely unexplored. Here, we use transverse focusing of electrons in a perpendicular magnetic field (TMF) (12, 19–23) to probe the properties of moiré minibands in TBG and demonstrate an exceptionally high quality of the “artificial metal” in TBG, as well as a possibility to use vertical displacement field,  $D$ , to break the valley degeneracy in the two constituent layers and selectively enhance transport in one of the minivalley.

## RESULTS

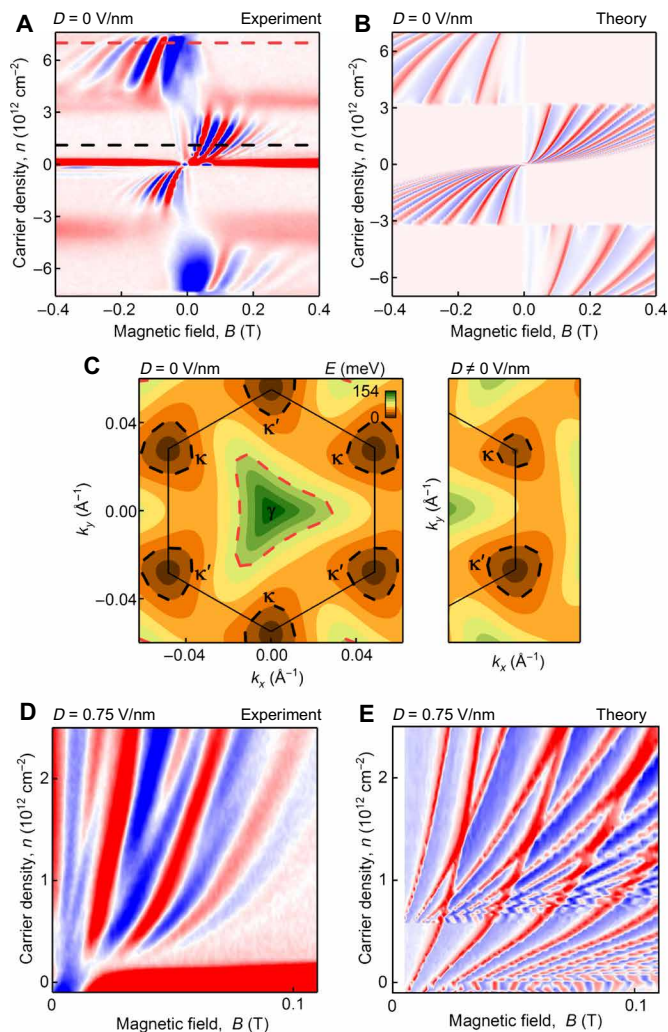
### Studied devices

We studied two high-quality dual-gated TBG devices encapsulated with  $\sim 30$ - to  $50$ -nm-thick hexagonal boron nitride (hBN) crystals: D1, with  $\theta = 1.87^\circ \pm 0.01^\circ$  (shown in Fig. 1C), and D2, with  $\theta = 2.60^\circ \pm 0.01^\circ$  (fig. S1A). The procedure used to determine  $\theta$  is described in section S1. The devices were fabricated using standard dry-transfer (24, 25) and tear-and-stack (4, 26) techniques (see section S2 for details). To ensure a clean interface between the two graphene layers, special care was taken to avoid any contact between graphene and the polymer during the transfer (section S2). In transport measurements, both devices showed similar behavior, with low-temperature mobilities in excess of  $400,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  for carrier density  $n \sim 10^{12} \text{ cm}^{-2}$ . All data shown below were obtained at a constant displacement field,  $D$ , that was achieved by a simultaneous sweep of the top and bottom gates (section S2).

### Transverse magnetic focusing

The high mobility for both devices enabled observation of TMF (12, 19–23), which is a manifestation of ballistic motion of electrons and had been used to characterize the shape of Fermi surfaces in both 3D (19, 20) and 2D (12, 21–23) metals. To measure the effect of TMF in our TBG devices, we used a nonlocal geometry illustrated in Fig. 1C, where narrow contacts 1 and 2 at one end of the device were used for current injection (driving current  $I_{12}$ ) and contacts 3 and 4 at the other end were used to detect a voltage  $V_{34}$ . In the presence of a perpendicular magnetic field, electrons injected from contact 1 propagate along the device edges in skipping orbits and form a characteristic caustic pattern determined by the shape of the Fermi surface, as illustrated in the insets of Fig. 1D. Caustics are focused into equidistant focal points along the sample edge, and the drift direction of the skipping orbits is determined by the sign of the magnetic field such that electron- and hole-like carriers propagate in opposite directions. As the positions of focal points vary with the magnetic field, whenever they coincide with the position of the voltage probe (contact 3 in Fig. 1C), one observes a focusing peak in the nonlocal resistance  $R_f = V_{34}/I_{12}$ . Figure 1D gives two examples of the observed focusing peaks measured at different carrier densities.

Figure 2A shows a typical dependence of  $R_f$  on the carrier density and magnetic field at zero displacement field,  $D = 0 \text{ V nm}^{-1}$ . Here, the appearance of an  $R_f$  signal in a particular quadrant of the  $B$ - $n$  diagram reflects the sign of the cyclotron mass, while the change of the quadrant upon doping indicates an inversion of the electron dispersion (i.e., a change of sign of the mass from electrons to holes or vice versa). Accordingly, a fan-like pattern in the center of Fig. 2A, which converges and changes direction at zero carrier density, indicates a neutrality point. Two additional, qualitatively similar, changes of the cyclotron mass appear at higher electron and hole densities,



**Fig. 2. Transverse magnetic focusing map.** (A) Focusing signal  $R_f$  as a function of the magnetic field and carrier density measured at 2 K for device D1 in zero displacement field,  $D = 0 \text{ V nm}^{-1}$ . Color scale: blue to red,  $\pm 3$  ohms. (B) TMF map calculated from the energy spectrum shown in Fig. 1B using a numerical method described in section S4. The angle between the zigzag edge of one of the monolayers and the sample boundary is taken as  $45^\circ$  to avoid any spurious effects of crystallographic alignment. As demonstrated in section S4, the calculated TMF map is only very weakly sensitive to the mutual orientation between graphene and the sample edge, confirming the generality of our results. (C) Contour plot of the first conduction miniband shown for the K valley of the Brillouin zone for zero (left) and nonzero (right) displacement fields. Black and red dashed lines outline the shape of the Fermi surfaces for carrier densities marked by black and red dashed lines in (A); the latter corresponds to equivalent doping levels relative to the main (black) and secondary (red) neutrality points. The color scale is from 0 to 154 meV. (D)  $R_f$  as a function of magnetic field and carrier density for device D2 measured at  $T = 2 \text{ K}$  and  $D = 0.75 \text{ V nm}^{-1}$  at a distance of  $8.5 \mu\text{m}$  from the injector (more data are shown in fig. S5). Color scale: blue to red,  $\pm 0.2$  ohm. (E) TMF map calculated numerically for device D2 in a displacement field (see sections S3 to S5 for details), which shows the splitting of the focusing peaks originating from the different miniband dispersion at  $\kappa$  and  $\kappa'$ .

showing inverted fan-like patterns at higher energies. These indicate that the electron dispersion converges toward a new (secondary) neutrality point, such as shown in Fig. 1B. The crossover between these two regimes (at  $n \approx 3 \times 10^{12} \text{ cm}^{-2}$  and  $-3 \times 10^{12} \text{ cm}^{-2}$ ) must

correspond to a van Hove singularity (vHS) in the moiré miniband spectrum. For a quantitative comparison, Fig. 2B presents the results of TMF modeling for device D1. Here, we used the model from (3) to compute the electron spectrum shown in Fig. 1B (see section S3 for details) and to perform numerical analysis of caustics (12), where the contributions to  $R_f$  from trajectories of electrons leaving the injection contact at different angles were weighted proportionally to  $|\nabla_k E|^{-1}$  (section S4). A good agreement between the experiment (Fig. 2A) and theory (Fig. 2B) suggests that the band structure of TBG is well described by the spectrum shown in Fig. 1B.

It is noticeable that the fan-like patterns in Fig. 2 (A and B)—corresponding to the main and secondary neutrality points of the TBG superlattice (around zero carrier density and above the vHS, respectively)—have different periodicities. This difference is caused by different sizes of the Fermi surfaces at equivalent doping levels (black and red dashed lines in Fig. 2A), due to the degeneracy of the miniband dispersion at  $\kappa$  and  $\kappa'$ . The Fermi surface contours are shown in Fig. 2C by black dashed lines around  $\kappa$  and  $\kappa'$  points of the mini Brillouin zone (main neutrality point) and a red dashed line around the  $\gamma$  point (secondary neutrality point). Furthermore, our theoretical analysis suggests that the Fermi surfaces close to the  $\gamma$  point have a triangular shape (Fig. 2C), which can be traced to the strong interlayer hybridization of those states. At the same time, the Fermi surfaces around  $\kappa$  and  $\kappa'$  points (that coincide with the valley centers K of the top and bottom graphene layers) are almost isotropic, as in monolayer graphene, pointing toward weak interlayer hybridization of these states.

### Effect of the displacement field

The absence of appreciable interlayer coupling at  $\kappa$  and  $\kappa'$  can be used to disentangle the TMF contributions from different minivalleys. To this end, we used a finite displacement field, up to  $D = 0.75 \text{ V nm}^{-1}$  (achievable without a risk of damaging our devices), which shifts the on-layer potential for electrons and therefore shifts the energies of the Dirac cones at  $\kappa$  and  $\kappa'$ , as illustrated in Fig. 2C. Such layer-symmetry breaking lifts the degeneracy between  $\kappa$  and  $\kappa'$  and separates the motion of electrons from different minivalleys in a magnetic field, as they now have different sizes of cyclotron orbits. This generates two different magneto-oscillation frequencies of  $R_f$  at low carrier den-

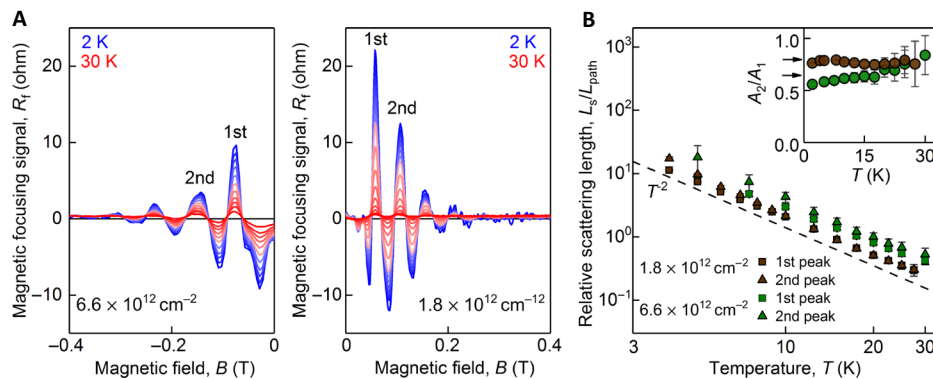
sities,  $|n| < 10^{12} \text{ cm}^{-2}$ , as seen in Fig. 2D, where separate focusing peaks appear for the electrons from each minivalley.

### DISCUSSION

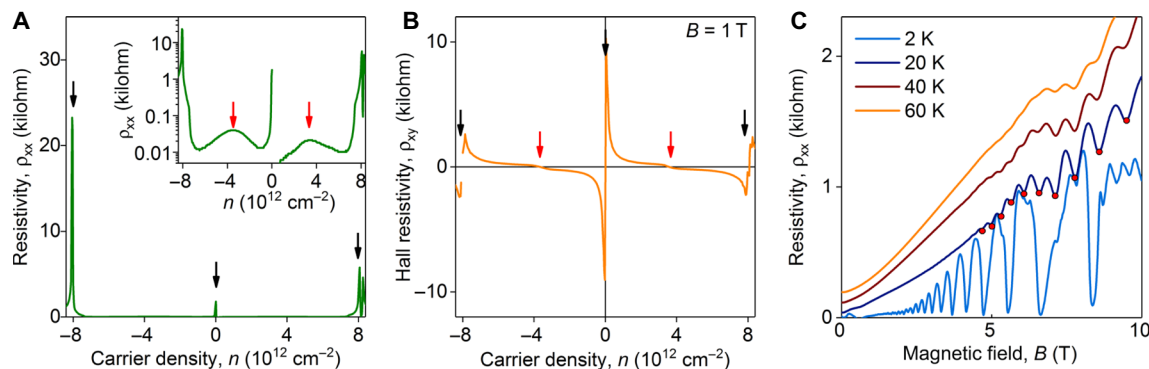
Further information about carrier dynamics in TBG can be obtained by studying the temperature dependence of TMF and its evolution for consecutive focusing peaks. In Fig. 3A, we show how the amplitude of TMF oscillations depends on temperature  $T$  in the range  $2 \text{ K} < T < 30 \text{ K}$ , in the vicinity of both main and secondary neutrality points. For quantitative analysis, we extract the relative scattering length as (12)

$$\frac{L_s}{L_{\text{path}}} = \left( \ln \left[ \frac{A(T_{\text{base}})}{A(T)} \right] \right)^{-1} \quad (1)$$

where  $L_{\text{path}}$  is the length of trajectories extending from the injector to the first focal point as shown in Fig. 1D, and  $A_{1(2)}(T)$  and  $A_{1(2)}(T_{\text{base}})$  are the areas under the first (second) focusing peak in Fig. 3A at  $T$  and  $T_{\text{base}} = 2 \text{ K}$ , respectively. The results are shown in Fig. 3B. The measured scattering lengths  $L_s$  for both carrier densities and all focusing peaks follow a  $T^{-2}$  scaling, which is different from the  $T^{-1}$  dependence characteristic of phonon-dominated scattering (23, 27). Such scaling points toward the dominance of low-angle electron-electron scattering that was also found to be responsible for the TMF suppression in graphene/hBN superlattices (12). Furthermore, the ratio between the areas under the second and first focusing peaks in Fig. 3A,  $A_2/A_1$ , characterizes the reflection of electrons at the sample boundary: The closer it is to one, the higher the probability for the incoming electrons to undergo specular reflection. In our experiment, electrons with energies near the main neutrality points ( $n \approx 1.8 \times 10^{12} \text{ cm}^{-2}$ ; Fig. 3A, right) undergo almost specular reflection ( $A_2/A_1 \approx 0.8$ ), while reflection of the electrons with energies near the secondary neutrality point ( $n \approx 6.6 \times 10^{12} \text{ cm}^{-2}$ ; Fig. 3A, left) is notably less specular ( $A_2/A_1 \approx 0.65$ ). This indicates a higher probability of diffusive scattering in the latter case, which is consistent with the greater sensitivity of the corresponding part of the miniband spectrum to inevitable perturbations of the moiré pattern near the sample edge. Because of little hybridization between the layers near  $\kappa$  and  $\kappa'$ , the scattering of Dirac electrons should be little affected by the termination of



**Fig. 3. Temperature dependence of magnetic focusing.** (A) Temperature dependence of the TMF signal measured at two characteristic carrier densities for device D1 (see legends).  $T$  was varied from 2 to 30 K (blue to red). (B)  $T$  dependence of the relative scattering length (see text) extracted from experimental data for consecutive focusing peaks. Absolute scattering lengths for several relative orientations of the crystallographic axes and the sample edge are shown in fig. S6. Dashed line shows  $T^{-2}$  dependence. The inset shows the ratio of the areas under the first and second focusing peaks in (A) as a function of  $T$ . Arrows correspond to  $A_2/A_1 = 0.8$  and  $0.65$  (see text). Error bars indicate the accuracy of determining  $A_2/A_1$ ; large errors at  $T > 20 \text{ K}$  are due to the relatively large background signal as the focusing peaks become strongly suppressed.



**Fig. 4. Bulk transport properties of TBG.** (A) Resistivity as a function of carrier density measured at 5 K for device D1. The inset shows the same data on a logarithmic scale. (B) Hall resistivity as a function of the carrier density for D1. Black arrows in (A) and (B) mark neutrality points, and red arrows mark vHS. (C) Resistivity versus magnetic field measured at different temperatures for device D1 at  $n = 3.1 \times 10^{12} \text{ cm}^{-2}$ . Red dots on a 40-K curve highlight the positions of Brown-Zak oscillations.

superlattice periodicity near the edge, while its part near the secondary neutrality points should be affected substantially, promoting diffusive scattering.

Last, we note that the above observations of superlattice effects in TMF correlate well with the bulk transport properties of the same TBG samples studied using local geometry. The longitudinal and Hall resistivity shown in Fig. 4 (A and B) displays secondary neutrality points (indicated by black arrows) and vHS (red arrows) at the same carrier densities as those inferred from the TMF experiments. Furthermore, the presence of a moiré superlattice in the studied TBG samples is seen from the presence of Brown-Zak oscillations (13, 14) that dominate the magnetotransport above  $T \sim 30 \text{ K}$  (Fig. 4C): While at low temperatures the magnetoresistance is dominated by Shubnikov-de Haas oscillations (see  $T = 2 \text{ K}$  curve in Fig. 4C), these are rapidly suppressed as  $T$  increases and give way to another  $1/B$ -periodic oscillations, with period determined by the relation between the magnetic flux through the moiré supercell area,  $A_S$ , and the magnetic flux quantum  $\phi_0 = h/e$ , i.e.,  $BA_S = \phi_0/q$  (where  $q$  is an integer).

To conclude, we have demonstrated that TBG supports ballistic propagation of electrons in multimicrometer devices, with electron transport determined by the reconstruction of the energy spectrum in the presence of a long-period superlattice. This offers new opportunities to study fundamental phenomena, such as Bloch oscillations in moiré superlattices (28–30) and their use for, e.g., terahertz generation. Moreover, we have shown that the sensitivity of the TBG band structure to the displacement field allows selective manipulation of electrons from different minivalleys, which may be implemented in electronic devices exploiting the valley degree of freedom.

## MATERIALS AND METHODS

### Device fabrication

To make TBG, we used the standard tear-and-stack method as described in section S2. TBG stacks were encapsulated between hBN crystals and assembled onto  $\text{SiO}_2$  substrate with doped silicon underneath, which served as a bottom gate electrode. Then, devices were shaped into hall bars using standard electron-beam lithography and reactive-ion etching techniques (section S2).

### Measurement details

We used standard low-frequency lock-in measurement technique with excitation frequency of 10 to 30 Hz. To independently control

the carrier density and interlayer displacement field, we used dual-gated geometry, as discussed in section S2.

## SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at <http://advances.sciencemag.org/cgi/content/full/6/16/eaay7838/DC1>

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