Research shows the 'magic range' of twisted bilayer graphene is larger than previously expected

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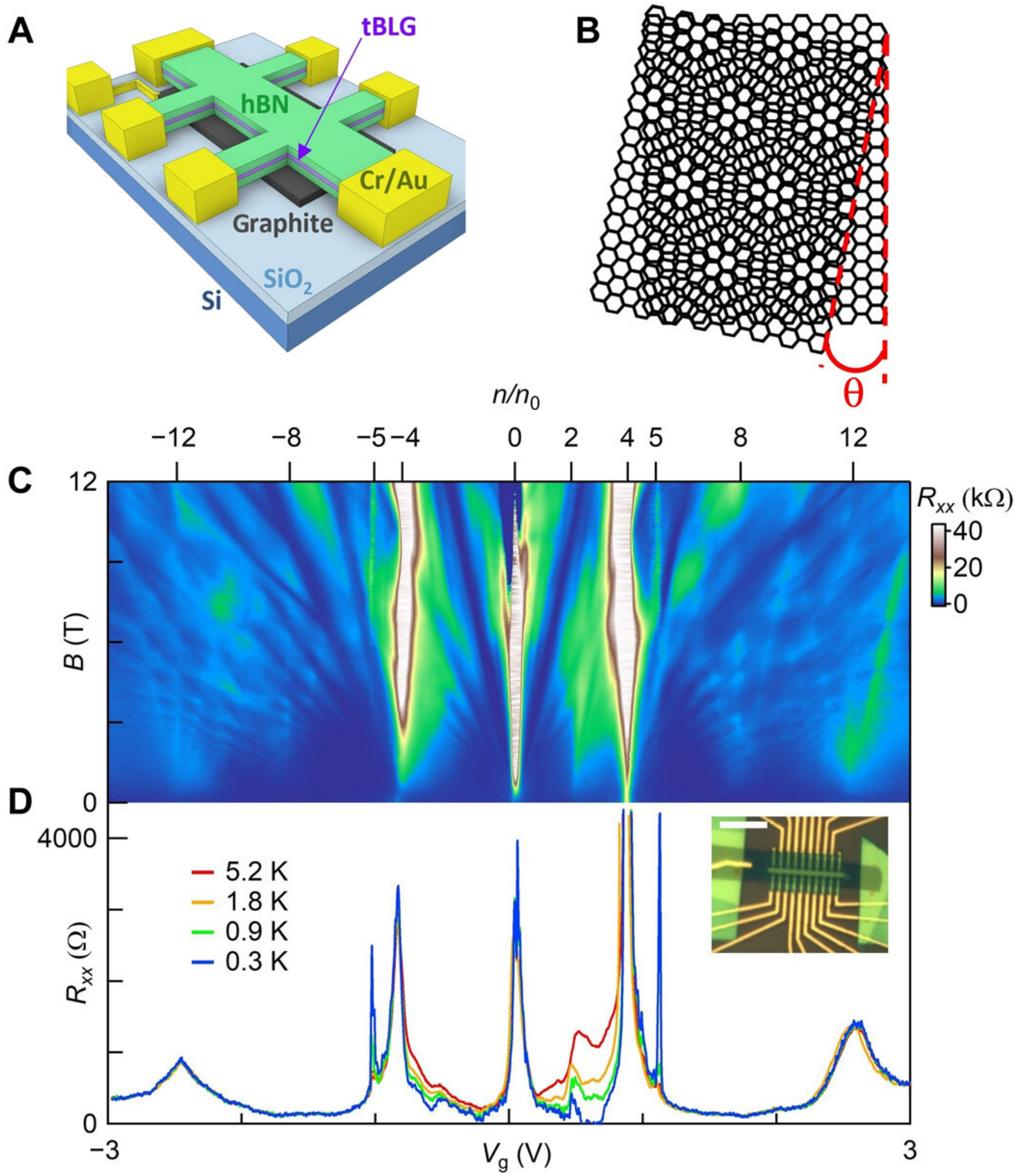
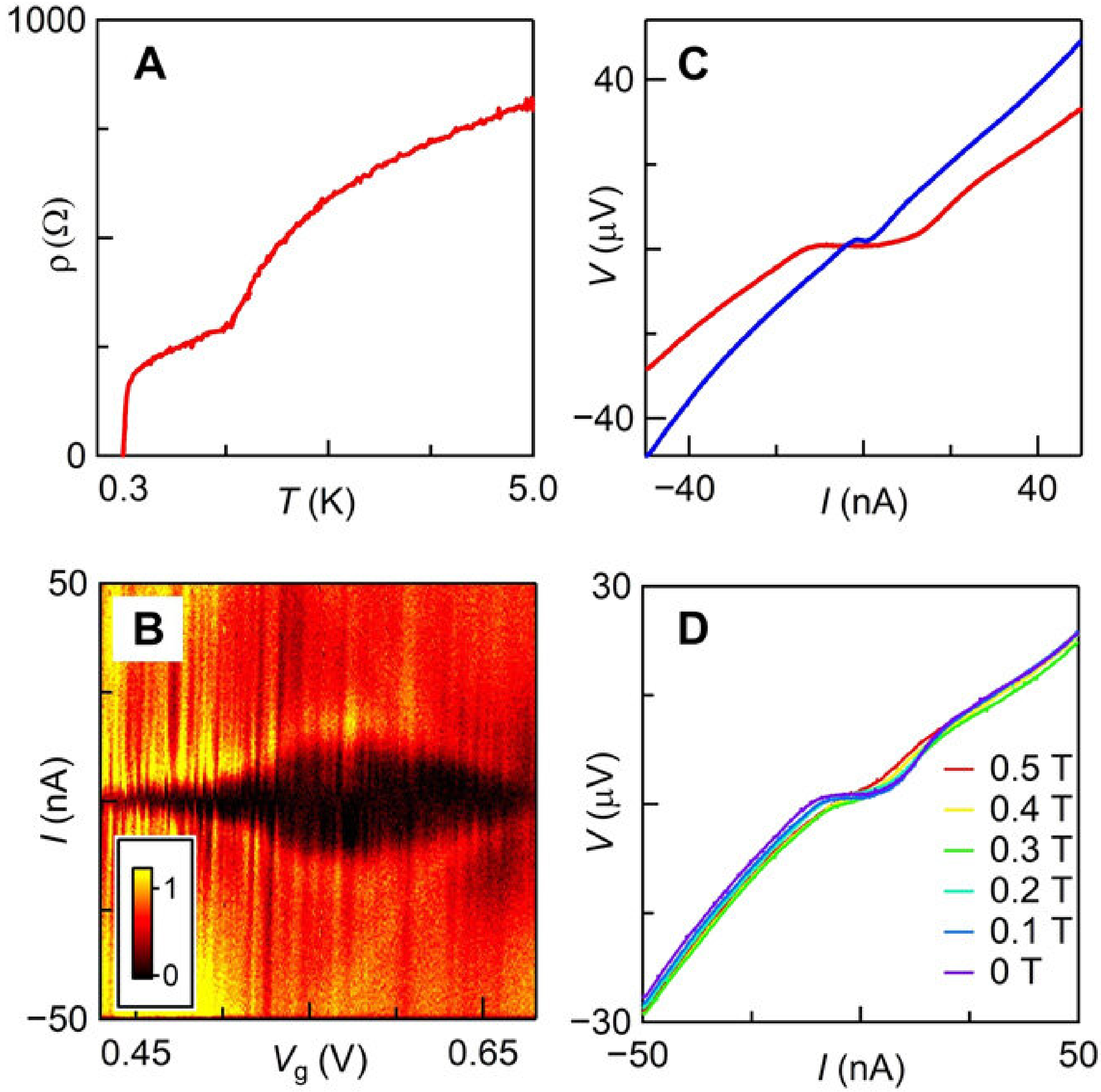


Device geometry and magneto-transport data. (A) Schematic diagram of device geometry. (B) Schematic diagram of moiré superlattice formed by the twisted graphene layers. (C) Rxx versus magnetic field B and gate voltage Vg showing a Landau fan pattern. The top axis labels nm, the number of charges per superlattice cell. (D). Rxx(Vg) at different temperatures. Inset: Optical image of a tBLG device with a scale bar of 10 μm. Credit: Science Advances, doi: 10.1126/sciadv.aaw9770

In materials science and quantum physics, flat bands and correlated behaviors within the "magic angle" twisted bilayer graphene (tBLG) has sparked significant interest, although many of its properties face intense debate. In a new report published in *Science Advances*, Emilio Codecido and colleagues in the departments of physics and materials science in the U.S. and Japan observed both superconductivity and a Mott-like insulator state in a tBLG device with a twist angle approximating 0.93 degrees. This angle was 15 percent smaller than the magic angle computed (~ 1.1°) in previous studies. The study revealed the "magic" range of tBLG to be larger than previously expected. The work provided a wealth of new information to decipher the strong quantum phenomena within tBLG devices for applications in quantum physics.

Physicists define 'Twistronics' as the relative twist angle between adjacent van der Waals layers to produce a moiré superlattice and flat bands in graphene. The concept has emerged as a new and uniquely suitable approach to markedly alter and tailor two-dimensional materials-based device properties to enable the flow of electricity. The marked effect of Twistronics is exemplified in groundbreaking recent work by researchers who demonstrated the emergence of extremely flat bands when two monolayer graphene layers were stacked at a magic twist angle of $\theta = 1.1 \pm 0.1°$.

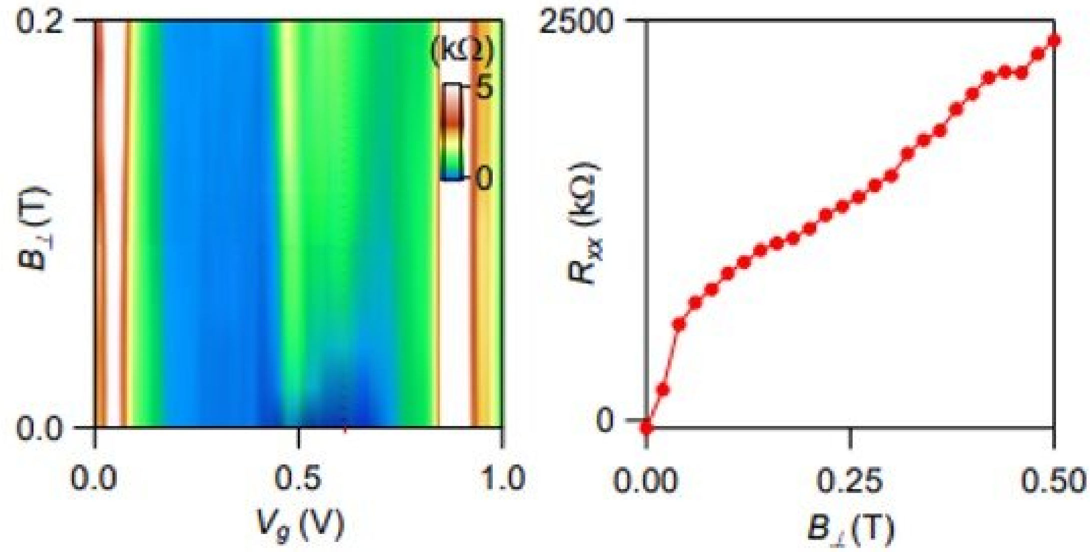
In the present work, Codecido et al. experimentally observed an insulting phase at half filling of the superlattice's first miniband (structural feature) in the twisted bilayer graphene (tBLG) device at the magic angle. The research team identified this to be a Mott insulator (an insulator with properties of superconductivity) exhibiting superconductivity at slightly higher and lower doping. The phase diagram revealed high-temperature superconductors between the superconductivity transition temperature (Tc) and the Fermi temperature (T_F). The work sparked tremendous interest and theoretical debate on the semiconductor system relative to the energy band structure, topology and additional magic angles of graphene. Compared to the initial theoretical reports, experimental studies are scarce and only just beginning to emerge.



Data from the superconducting state. (A) ρ versus temperature when the density is tuned to the superconducting phase (Vg ~ 0.53 V or nm ~ 2.5). (B) Differential resistance dV/dI versus bias current and gate in the superconducting phase at base temperature (280 mK). Color scale is in units of kilohms. (C) Voltage-current characteristics at T = 280 mK and Vg = 0.50 V (blue) and 0.58 V (red), respectively. (D) V-I curves at different parallel magnetic fields. Credit: Science Advances, doi: 10.1126/sciadv.aaw9770

In this study, the research team conducted transport measurements of a magic angle tBLG device exhibiting correlated insulating and superconducting states. They unexpectedly obtained a twist angle at 0.93 ± 0.01, which was 15 percent smaller than the magic angle already established, while being the smallest reported to date and exhibiting superconductivity. These results indicated that the new correlated states could emerge in the tBLG device below the primary magic angle and beyond the first miniband of graphene.

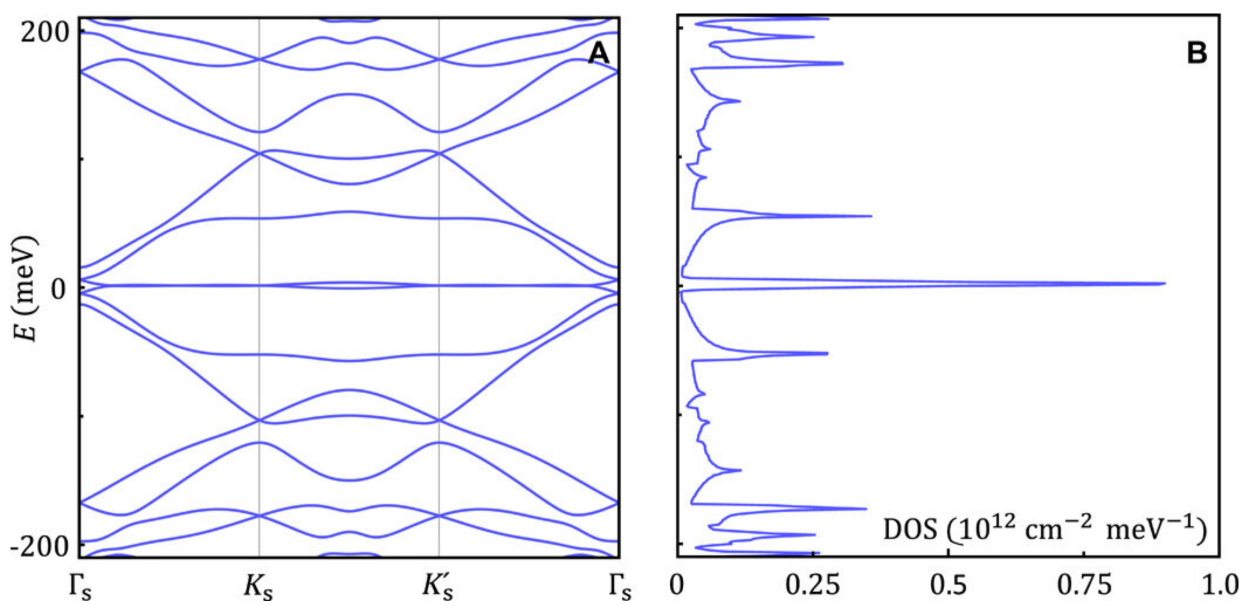
To build the devices, the research team used the "tear and stack" approach. They encapsulated the construct between hexagonal boron nitride (BN) layers; patterned into a Hall bar geometry with multiple leads coupled to Cr/Au (chromium/gold) edge contacts. They fabricated the entire device on top of a graphene layer that served as the back gate. Codecido et al. measured the devices in pumped He^4 and He^3 cryostats using standard direct current (DC) and alternate current (AC) lock-in techniques. The team recorded the device's longitudinal resistance (Rxx) vs. an extended gate voltage (Vg) range and calculated the magnetic field B at a temperature of 1.7 K. They observed the small electron-hole asymmetry to be intrinsic to tBLG devices as observed in previous reports. The team noted the results to detail the smallest twist angle value reported to date for tBLG devices exhibiting superconductivity.



Superconductivity response to magnetic field. Rxx (Vg, B⊥) illustrating the disappearance of superconductivity with perpendicular magnetic field. Credit: Science Advances, doi: 10.1126/sciadv.aaw9770

On closer examination of the Landau fan diagram, Codecido et al. obtained a number of salient features. For instance, the peak at half filling and the two-fold degeneracy of Landau levels were consistent with previous observations of a Mott-like correlated insulating state. The team showed breaking of approximate spin-valley SU (4) symmetry and the formation of a new quasi-particle Fermi surface. However, the details required more delicate examination. They also observed the emergence of superconductivity, which increased the Rxx (longitudinal resistance), similar to preceding work.

The team then investigated the critical temperature (Tc) of the superconducting phase. Since the data was not obtained at optimal doping for superconductivity in this sample, the scientists assumed Tc could be as high as 0.5 K. However, the device became nonfunctional before they could obtain clear data from the superconducting state. To further investigate the superconducting state, they measured the four-terminal voltage-current (V-I) characteristics of the device at different carrier densities. They obtained displays of resistance and observed the supercurrent for an extended range of density and showed the suppression of supercurrent on application of a parallel magnetic field. To gain insight to the behavior observed in the study, Codecido et al. calculated the moiré band structure for the tBLG device using the Bistritzer-MacDonald model with refined parameters.



Calculations of electronic band structures of 0.93° tBLG. (A) Energy dispersion. (B) Density of states (DOS). In obtaining the DOS from the band structure, 1 meV was used for the energy interval, and the spin-valley degeneracy was considered. Credit: Science Advances, doi: 10.1126/sciadv.aaw9770

In contrast to previous calculations of the magic angle, the research team showed the calculated low-energy moiré Dirac bands were not as energetically isolated from the high-energy bands. Although the twist angle of the device was smaller than the magic angle computed elsewhere, the device hosted phenomena (Mott-like insulating and superconductivity) that strongly correlated with previous studies. The physicists found this to be both unexpected and desirable.

On further evaluation of behavior at large density (the number of available states at each energy) the scientists credited the observed features to a newly emerging correlated insulating state. They propose additional delicate studies of the density of states (DOS) in the future to understand the exotic insulating states and determine if or not they can be classified as quantum spin liquids.

In this way, Emilio Codecido and colleagues observed superconductivity near a Mott-like insulating state within a twisted bilayer device at a small twist angle (0.93°). The work showed the influence of electron correlations on the properties of moiré superlattices even at such small angles and high densities. Future work will investigate the spin-valley ordering of the insulating phases and investigations at lower temperatures in their search for new superconducting phases. The experimental studies will be coupled with theoretical efforts to understand the origins of this behavior.

More information: Emilio Codecido et al. Correlated insulating and superconducting states in twisted bilayer graphene below the magic angle, *Science Advances* (2019). DOI: 10.1126/sciadv.aaw9770

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