**2D MATERIALS**

**Tuning electron correlation in magic-angle twisted bilayer graphene using Coulomb screening**

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Controlling the strength of interactions is essential for studying quantum phenomena emerging in systems of correlated fermions. We introduce a device geometry whereby magic-angle twisted bilayer graphene is placed in close proximity to a Bernal bilayer graphene, separated by a 3-nanometer-thick barrier. By using charge screening from the Bernal bilayer, the strength of electron-electron Coulomb interaction within the twisted bilayer can be continuously tuned. Transport measurements show that tuning Coulomb screening has opposite effects on the insulating and superconducting states: As Coulomb interaction is weakened by screening, the insulating states become less robust, whereas the stability of superconductivity at the optimal doping is enhanced. The results provide important constraints on theoretical models for understanding the mechanism of superconductivity in magic-angle twisted bilayer graphene.

The discovery of superconductivity in magic-angle twisted bilayer graphene (tBLG) has raised intriguing questions about the nature of the superconducting order parameter (7–9). The phase diagram of magic-angle tBLG, featuring both the correlated insulator (CI) and superconducting phases, resembles that of cuprate materials, suggesting that the superconducting phase arises from an unconventional origin (1, 4–6, 7–12). By contrast, the more recent observations of superconductivity in the absence of CI appear to indicate that superconductivity arises through electron-phonon coupling (13–15), an interpretation that is backed by a range of theoretical models (16–18).

It has long been recognized that elucidating the role of Coulomb interaction is essential for determining the nature of superconductivity. For a conventional superconductor, electron-phonon coupling competes against Coulomb repulsion in stabilizing superconductivity at low temperature (19, 20). As such, a weaker Coulomb repulsion will lead to more robust superconducting order parameters. By contrast, an unconventional superconducting phase arises from an all-electron mechanism, whereby the order parameter strengthens with increasing Coulomb interaction (4, 5). For conventional solid-state materials, it remains an experimental challenge to directly control Coulomb interaction within a superconductor without introducing additional changes to the material. The flexibility of the van der Waals materials offers a valuable opportunity to control Coulomb interaction in magic-angle tBLG structure using proximity screening (13, 14, 21).

A major roadblock for addressing the mechanism underlying the superconducting phase is the vastly different behavior across different systems of correlated fermions. We introduce a device geometry whereby magic-angle twisted bilayer graphene is placed in close proximity to a Bernal bilayer graphene, separated by a 3-nanometer-thick barrier. By using charge screening from the Bernal bilayer, the strength of electron-electron Coulomb interaction within the twisted bilayer can be continuously tuned. Transport measurements show that tuning Coulomb screening has opposite effects on the insulating and superconducting states: As Coulomb interaction is weakened by screening, the insulating states become less robust, whereas the stability of superconductivity at the optimal doping is enhanced. The results provide important constraints on theoretical models for understanding the mechanism of superconductivity in magic-angle twisted bilayer graphene.

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**Fig. 1. Hybrid double-layer structure with Bernal BLG and tBLG.** (A) Schematic of the hybrid double-layer structure consisting of a Bernal BLG and a magic-angle tBLG, separated by a thin insulating barrier with thickness of $t = 3$ nm. The structure is encapsulated with dual hexagonal boron nitride (hBN) dielectric and graphite gate electrodes. (B) Longitudinal resistance of Bernal BLG $R_{BLG}$ as a function of $D_{BLG}$ and $n_{BLG}$ at $T = 20$ mK. (C) Longitudinal resistance of tBLG $R_{tBLG}$ as a function of $V_{top}$ and $V_{bot}$, measured at $T = 20$ mK and $V_{bot} = -200$ mV. Screening from BLG is minimal between the white dashed lines, where BLG is fully insulating and tBLG is tuned with both top and bottom graphite gates, giving rise to the distortion in transport features. Inset: Schematic energy structure of BLG at large $D_{BLG}$ for three values of $n_{BLG}$. (D) $R_{tBLG}$ as a function of filling fraction in tBLG $n_{tBLG}$, measured at $D_{tBLG} = -350$ mV/\(\text{nm}\) and $n_{tBLG} = 0$ with varying temperature.

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Gxx to screen the Coulomb interaction in the bottom graphite gates, n and tBLG samples near the magic angle, which may Fig. 2. The effect of Coulomb screening on the CI. (A) Arrhenius plot for the correlated insulating state at $n_{BLG} = -2$, measured at $D_{BLG} = -400$ mV/nm with varying carrier density $n_{BLG}$. Four-terminal conductances $G_{xx}$ measured at different $n_{BLG}$ are normalized by their value at $T = 5$ K. (B) Activation energy gap of the $n_{BLG} = -2$ CI state as a function of $n_{BLG}$.

tBLG samples near the magic angle, which may result from the spatial inhomogeneity of the moiré pattern (22–24). The variation between different samples makes it difficult to provide reliable experimental constraints on a theoretical model. Here, we address this obstacle by tuning the strength of the Coulomb interaction in a single device using screening, while studying the response in both the CIs and the superconducting phase using transport measurement. We use a hybrid double-layer structure, in which a Bernal bilayer graphene (BLG) and a magic-angle twisted bilayer graphene (tBLG) are separated by a thin insulating barrier with a $\delta = 3$ nm thickness (Fig. 1A). The close proximity allows charge carriers from BLG to screen the Coulomb interaction in the tBLG, offering direct control of electron correlations in the moiré flat band.

Charge carrier density in BLG and tBLG, $n_{BLG}$ and $n_{tBLG}$, can be independently controlled by varying the applied voltage to the top and bottom graphite gates, $V_{top}$ and $V_{bot}$ (25). Additionally, a voltage bias across two layers $V_{int}$ induces a perpendicular electric field, $D$, providing experimental control for the energy gap at the charge neutrality point in BLG (26–28). Longitudinal resistance measured from the Bernal BLG displays a well-defined peak at the neutrality point, which grows more insulating with increasing $D$-field (Fig. 1B). At large $D$-field, Bernal BLG can be tuned from fully insulating at $n_{BLG} = 0$ (blue circle in Fig. 1B) to highly conductive at large $n_{tBLG}$ (red circle in Fig. 1B), offering a large contrast in the strength of Coulomb screening. Figure 1C plots the transport response of tBLG with a twist angle of $\theta = 1.04^\circ$ (25), as $n_{BLG}$ and $n_{tBLG}$ are both tuned by varying $V_{top}$ and $V_{bot}$. A normalized density scale, marked by red arrows, is defined to describe partial filling $v$ of the moiré band on the basis of the fourfold degeneracy of spin and valley degrees of freedom for both electron- and hole-type carriers. The static $\varepsilon/\mathbf{r}$ Coulomb interaction among the electrons with charge $e$, separated by distance $r$ in the tBLG, is modified by the presence of BLG, leading to an effective interaction energy $V^{\text{eff}}(r) = \frac{e^2}{(2\pi \varepsilon_0)^2} \left[1 - e^{-2\pi q r} \left(1 - \frac{1}{\varepsilon_{AB}(q)}\right)\right] e^{i q r}$

The wave vector $\mathbf{q}$-dependent dielectric constant of the BLG can be related to its static polarization function $\Pi_0^{\text{eff}}(q)$ as $\varepsilon_{AB}(q) = 1 + \frac{2\pi}{q^2} \Pi_0^{\text{eff}}(q)$. When the BLG is gated to a finite carrier density and thus acts as a metal, $\Pi_0^{\text{eff}}(q) \rightarrow \text{const.}$ and the $\varepsilon_{AB}(q \rightarrow 0)$ diverges. At long distances, $V^{\text{eff}}(r)$ then corresponds to the real space potential produced by the test charge and its mirror image a distance $2\delta$ above the twisted bilayer. When the BLG is insulating, $\Pi_0^{\text{eff}}(q) \sim q^2$ and the $V^{\text{eff}}(r)$ is unchanged at long distances by the presence of the BLG [see (25) for details of Coulomb screening in a hybrid double-layer structure at any $r$]. Because the strength of Coulomb screening is correlated with the conductivity of BLG, its effect can be studied by comparing transport properties of tBLG in and outside the density range marked by the white dashed lines in Fig. 1C.

First, we examine the transport response of tBLG in the absence of Coulomb screening, by measuring inside the white dashed lines in Fig. 1C, where BLG is fully insulating. Figure 1D plots the longitudinal resistance of tBLG, $R_{BLG}$, over the full filling range of the moiré flat band. Apart from the charge neutral point (CNP), a series of resistive features emerge at $v = 2, +1,$ and $+3$, which are consistent with the CI states from previous observation (1–3). In addition, a robust superconducting phase emerges at low temperature, evidenced by $R_{BLG}$ dropping to zero. The robust CI and superconducting states establish an excellent starting point, allowing us to quantitatively examine the effect of Coulomb screening by studying the variation in transport behavior while varying $n_{BLG}$.

Figure 2A plots the temperature dependence of four-terminal conductance of the CIs at $v = -2$, which exhibits thermally activated behavior with strong $n_{BLG}$ dependence. An energy gap $\Delta$ can be extracted from the slope of the Arrhenius plot (dashed lines in Fig. 2A), providing a measure for the strength of the CI state. The effect of Coulomb screening is investigated by measuring the energy gap $\Delta$ while varying density in BLG $n_{BLG}$ (Fig. 2B). $\Delta_{v=-2}$ is largest when BLG is fully insulating at $n_{BLG} = 0$. Similar behavior in the energy gap is observed at $v = 2$ and 3 as a function of $n_{BLG}$ [see figs. S11 and S12 (25)]. Because CIs at integer fillings arise from Coulomb correlation within the moiré flat band (6, 29, 30, 31), the trend in $\Delta$ provides strong evidence that electron correlations in tBLG are directly tunable using Coulomb screening. Screening from the BLG decreases as BLG becomes insulating, leading to stronger Coulomb interactions and a larger $\Delta$ for the CI states. In addition, a minimum in $\Delta_{v=-2}$ is observed near the band edge of BLG at $n_{BLG} \sim 5 \times 10^{11} \text{ cm}^{-2}$, suggesting that Coulomb screening is strongest when the Fermi level of BLG is near the van Hove singularities, where the density of states is largest (32, 33). We note that BLG at $n_{BLG} = 0$ remains insulating over the temperature range of the thermal activation measurement, ensuring that the strength of Coulomb screening remains constant with varying temperature and is controlled only by $n_{BLG}$ [see figs. S13 and S14 (25)]. The robustness of the CI at $v = -2$ in the presence of strong Coulomb screening suggests that CIs cannot be fully suppressed by Coulomb screening alone (13). The effect of Coulomb screening on CIs shows excellent agreement with theoretical models (25), which provides an important reference for studying the nature of superconductivity in tBLG using Coulomb screening from BLG.

Having established the effect of Coulomb screening on CIs, we turn our attention to the superconducting phase on the hole-doping side of $v = -2$. Figure 3A plots $R_{BLG}$ at $T = 20$ mK as a function of carrier density in tBLG, $n_{tBLG}$. The superconducting phase is stable over a wider density range in the regime where BLG is metallic (red trace in Fig. 3A), as compared to when BLG is fully insulating (blue trace in Fig. 3A). The boundary of the superconducting region is defined by the density where $R_{BLG}$
Fig. 3. The effect of tuning $n_{BLG}$ in the presence of a large D-field–induced energy gap in BLG. (A) $R_{BLG}$ as a function of carrier density in tBLG. $n_{BLG}$, measured at $D_{BLG} = -350$ mV/nm with different $n_{BLG}$ at $T = 20$ mK. (B and C) $R_{BLG}$ as a function of carrier density $n_{BLG}$ and perpendicular magnetic field $B$ at $T = 20$ mK. BLG is tuned to $D_{BLG} = -350$ mV/nm with (B) $n_{BLG} = 0$ and (C) $n_{BLG} = -0.5 \times 10^{12}$ cm$^{-2}$. (D) Differential resistance $dV_x/dI$ versus d.c. bias current $I_{DC}$ measured at optimal doping $n_{BLG} = -1.48 \times 10^{12}$ cm$^{-2}$ and base temperature of $T = 20$ mK, with $D_{BLG} = 350$ mV/nm and BLG tuned to different density. A critical current $I_c$ is defined by the peak in $dV_x/dI$, where superconductivity is destroyed and the differential resistance goes over to the normal state value ($40$). (E) $R_{BLG}$ as a function of temperature measured at optimal doping $n_{BLG} = -1.48 \times 10^{12}$ cm$^{-2}$ and $D_{BLG} = -350$ mV/nm for different $n_{BLG}$. The blue and red traces are measured at $B = 0$, whereas the black trace is measured at the perpendicular magnetic field $B = 0.1$ T where superconductivity is fully suppressed. Inset: Temperature dependence of $R_{BLG}$ on a linear scale. $T_c$ operational definition is $50\%$ of normal state resistance and marked by the blue horizontal arrowhead. The separation of $B = 0$ and $B = 0.1$ T curves marks the onset of pairing (black vertical arrow).

electrical measurements of the critical current $I_c$, critical temperature $T_c$, and critical temperature $T_D$. The superconducting phase is more robust when BLG is metallic as compared to fully insulating. We note that $R_{BLG}$ measured at $B = 0.1$ T, where superconductivity is fully suppressed, reflects the transport behavior of tBLG in the normal state. The transition temperature $T_c$ operationally defined as $50\%$ of extrapolated normal state resistance, is shown to be $\sim 2.2$ K in Fig. 3E, which is in line with previous observations in tBLG with a similar twist angle ($14$). The temperature dependence of $R_{BLG}$ leads to a few important observations: (i) The normal state ($T \geq 3$ K) resistance in tBLG is insensitive to changes in BLG, demonstrating that modifications in impurity scattering resulting from a nearby metallic layer do not play a dominating role ($34$); (ii) the onset of Cooper pairing is observed at $T \sim 3$ K (vertical black arrow in Fig. 3E), evidenced by the bifurcation between $R_{BLG}$ measured at $B = 0$ and $0.1$ T. We note that the $R_{BLG}$-tuning and Cooper pairing onset at a similar temperature, as shown by the red and blue traces in Fig. 3E. The stability of the superconducting phase can be further explored by plotting $I_{DC}, T_c$, and $\Delta n_{SC}$ as a function of $n_{BLG}$. The values of all three properties closely follow the conductance of BLG, as shown in Fig. 4. A to D, with the superconducting phase at the optimal doping being more robust when BLG is metallic. Taken together, our measurements suggest that $n_{BLG}$ tuning has a direct impact on the stability of the superconducting phase in tBLG.

The enhancement of superconductivity by a nearby metallic BLG layer could, in principle, result from suppressed phase fluctuations in tBLG, without an appreciable effect on Cooper pair formation, giving rise to a higher Berezinskii-Kosterlitz-Thouless transition temperature $T_{BKT}$ ($35$). We can rule out this scenario because the $n_{BLG}$ tuning onsets at roughly the same temperature as Cooper pair formation.
Fig. 4. The effect of tuning $n_{BLG}$ on the superconductivity at the optimal doping in magic-angle tBLG. (A) The longitudinal conductance $G_{xx}$ in BLG as a function of carrier density $n_{BLG}$ measured at $D_{BLG} = -350$ mV/nm and $T = 20$ mK. Insets show the position of Fermi level relative to the energy band structure in BLG at different carrier densities. (B) $I_0$, (C) $T_c$, and (D) $\Delta T_{c}$ as a function of carrier density in BLG. $n_{BLG}$, measured at $D_{BLG} = -350$ mV/nm. $I_0$ and $T_c$ are measured at optimal doping $n_{BLG} = -1.48 \times 10^{12}$ cm$^{-2}$, $I_0$ and $\Delta T_{c}$ are measured at $T = 20$ mK. A large energy gap is induced in BLG by $D$-field. (E) $R_{x,x}$ as a function of temperature measured at optimal doping $n_{BLG} = -1.48 \times 10^{12}$ cm$^{-2}$ and $D_{BLG} = 0$ for different $n_{BLG}$. The blue and red traces are measured at $B = 0$, whereas the black trace is measured at $B = 0.1$ T, where superconductivity is fully suppressed. Tuning $n_{BLG}$ gives rise to small differences in $R_{xx}$ at temperatures much lower than $T_c$. No effect is observed in the temperature range near $T_c$. (F) $R_{xx}$ in BLG as a function of carrier density $n_{BLG}$ measured at $D_{BLG} = 0$ and $T = 20$ mK. (G) $T_c$ as a function of carrier density in BLG. $n_{BLG}$, measured at $D_{BLG} = 0$ and optimal doping $n_{BLG} = -1.48 \times 10^{12}$ cm$^{-2}$.

The in-plane superconducting phase in tBLG is strongly influenced by the Fermi level, which is controlled by the carrier density. The superconducting transition temperature $T_c$ increases with increasing carrier density. However, this increase is small compared to the change in the superconducting gap $\Delta T_{c}$, which decreases with increasing carrier density.

The superconducting gap $\Delta T_{c}$ is determined by the strength of the electron-phonon coupling, which is influenced by the Fermi level. The Fermi level is controlled by the carrier density, which is manipulated by the tuning of $n_{BLG}$. The results suggest that tuning $n_{BLG}$ can be used to control the superconducting properties of tBLG.

References and Notes

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**Competing interests:** The authors declare no competing financial interests.

**Data and materials availability:** Experimental data files are available at the Open Science Framework (41).

**SUPPLEMENTARY MATERIALS**

[Link to supplementary materials]

**References (44–44)**

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