

Tunable bandwidths and gaps in twisted double bilayer graphene on the verge of correlations

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We use temperature-dependent resistivity in small-angle twisted double bilayer graphene to measure bandwidths and gaps of the bands. This electron-hole asymmetric system has one set of nondispersing bands that splits into two flat bands with the electric field—distinct from the twisted bilayer system. With electric field, the gap between two emergent flat bands increases monotonically and bandwidth is tuned from 1 to 15 meV. These two flat bands with gap result in a series of thermally induced insulator to metal transitions—we use a model, at charge neutrality, to measure the bandwidth using only transport measurements. Having two flat bands with tunable gap and bandwidth offers an opportunity to probe the emergence of correlations.

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I. INTRODUCTION

The ability to tune the twist angle between two sheets of two-dimensional materials has enormously increased the scope of engineering superlattices [1–3]. Following theoretical predictions [4–6], some recent experiments have confirmed flat bands for a twisted bilayer graphene system near a magic angle. In addition, for some parameter space, it hosts highly correlated states showing exotic phenomena including a Mott insulator, superconductivity, and ferromagnetism [7–11]. The ratio of electron-electron interaction strength and width of the electronic band is a key parameter determining the emergence of correlations. It is desirable to find additional knobs to tune the bandwidth [12–14].

Twisted double bilayer graphene (TDBG) is also an interesting platform where one puts a copy of bilayer graphene on top of another one with a twist angle between them. The Bernal-stacked bilayer graphene has a unique band structure that changes upon applying perpendicular electric field as a band gap opens up with increasing field [15–18]. The TDBG system inherits this tunability with the electric field and makes itself an interesting system with electrically tunable flat bands [19–25]. However, there are only few experimental reports so far on this novel system where electric field tunable correlated insulating states and the realization of superconductivity have been demonstrated [26–29]. There is an urgent need to understand the single-particle band structure and this requires a detailed study of electron transport in a small-angle TDBG system to understand band gaps, bandwidths, and their tunability with the electric field.

In this work we present a comprehensive study of temperature dependence of electronic transport in a twisted double bilayer graphene device with a twist angle of 1.18° between the two sheets of bilayer graphene. This electron-hole asymmetric system has one set of low-energy nondispersing bands separated from higher energy dispersing bands by moiré gaps at zero perpendicular electric field. As we increase the electric field more, two clear trends emerge—first, after a finite electric field a band gap opens up at the charge neutrality point giving rise to two flat bands; this differentiates it from twisted bilayer graphene, where the low energy bands always touch at the Dirac points [19,21]. Second, further increase of the electric field closes the moiré gaps gradually in an asymmetric manner for the electron and hole side. By a systematic measurement of gaps as a function of electric field we establish both these trends experimentally. We then focus on understanding the thermally induced insulator to metal transitions in the parameter space of electric field and charge density. We show that this physics can be understood using a simple model with two flat bands at a finite perpendicular electric field. Applying this simple double quantum well with flat band model [30] we extract, using only transport measurements, the bandwidth tunable from ~ 1 to 15 meV by increasing the electric displacement field strength D/ϵ_0 from -0.24 to -0.34 V/nm. The moiré gaps separating the flat bands from the dispersing higher bands are ~ 10 meV. The fact that the gaps and bandwidths are tunable with the electric field makes this system important for studying correlations.

II. RESULTS AND DISCUSSIONS

We now discuss the details of our experiment. As shown in the schematic in Fig. 1(a), the twisted double bilayer graphene is encapsulated by both top and bottom few layers thick hexagonal boron nitride [3,31,32]. For details of the fabrication process, see Sec. I in the Supplemental Material [33]. The

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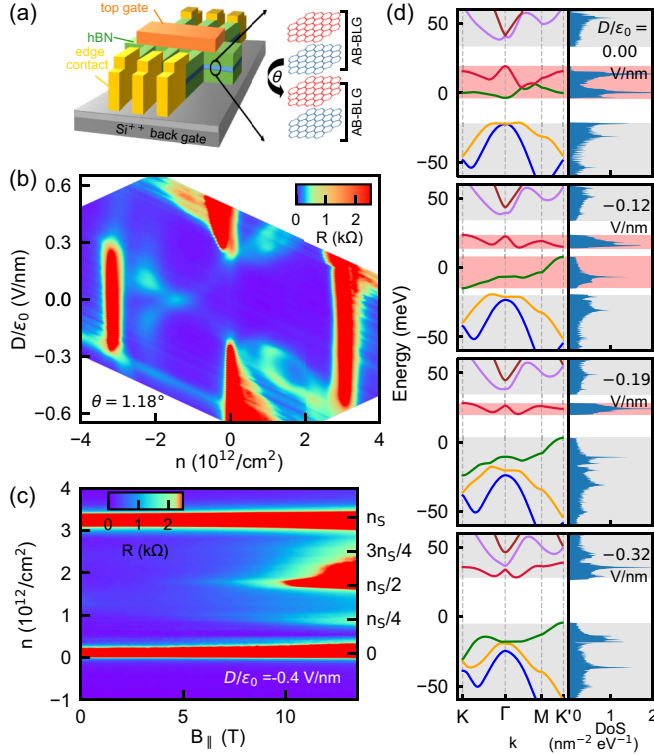


FIG. 1. The TDBG device, resistance variation, and DoS distribution. (a) The schematic of the dual gated twisted double bilayer graphene device. (b) Two-dimensional color scale plot of four-probe resistance as a function of carrier density (n) and the perpendicular electric displacement field (D). Gaps are formed for $n = 0$ and $\pm n_s$. (c) Development of correlation induced gaps at $D/\epsilon_0 = -0.4$ V/nm with the in-plane magnetic field up to 13.5 T. (d) The calculated low energy band structure (left panel) and corresponding density of states (right panel) at $B_{||} = 0$ illustrating the sequence of opening and closing of different gaps qualitatively. The energy range of the flat bands and the dispersing bands are shaded in pink and gray, respectively.

metallic top gate and the Si⁺⁺ back gate allow us independent control of both the carrier density (n) and the perpendicular electric displacement field (D), given by the equations $n = (C_{TG}V_{TG} + C_{BG}V_{BG})/e$ and $D = (C_{TG}V_{TG} - C_{BG}V_{BG})/2$, where C_{TG} and C_{BG} are the capacitance per unit area of the top and the back gate, respectively, e being the charge of an electron.

Figure 1(b) shows a color scale plot of the four-probe resistance R as a function of n and D measured at a temperature of 1.5 K. First, this TDBG device shows two pronounced peaks in resistance corresponding to the two moiré gaps at $n = \pm n_s$, where $n_s = 3.2 \times 10^{12} \text{ cm}^{-2}$ is the required charge density to fill the first moiré band corresponding to a twist angle of 1.18° . Another peak in resistance appears at the charge neutrality point (CNP) as the magnitude of D is increased indicating a gap opening with the electric field. This is also seen in a simple bilayer graphene device [16,17]. However, the resistance at the two moiré gaps decreases with increasing electric field, indicating a gradual closing of these gaps. Second, as reported in some very recent works on TDBG [26–29] there are several interesting features at specific parameter space,

namely, a crosslike feature around zero electric field only on the hole side and two halo regions of high resistance present at the electric displacement field $D/\epsilon_0 \sim \pm 0.4$ V/nm on the electron side. In this device we do not observe fully formed insulating states at filling other than $n = \pm n_s$ and $n = 0$ at zero magnetic field. However, Fig. 1(c) shows that with the in-plane magnetic field, gaps open up at $n_s/4$, $n_s/2$, and $3n_s/4$ for this device—suggesting that the system is on the verge of developing correlations. All of these observations indicate the existence of flat band physics and offer an opportunity to study the temperature dependence of transport properties in a flat band system. See Fig. S8 in the Supplemental Material [33] for data from another TDBG device with a twist angle 2.05° that shows two moiré gaps at $n = \pm n_s$ and gap opening at CNP, but no other features like the cross or the halo. This emphasizes the role of the flat bands in the device with 1.18° twist angle.

From Fig. 1(b) it is evident that this system possesses a high degree of electron-hole asymmetry. While the asymmetric presence of above mentioned crosslike feature and the halo feature are such examples, the different closing sequence of two moiré gaps offers another one. Different theoretical aspects of twisted angle systems have been understood using a minimal model where one sets further neighbor hoppings across the bilayer γ_3 and γ_4 , as well as the potential on dimer sites Δ' to be zero. However, to capture the electron-hole (e-h) asymmetry we use an e-h asymmetric model with $\gamma_3 = 320$ meV, $\gamma_4 = 44$ meV, and $\Delta' = 50$ meV (see Sec. II in the Supplemental Material [33] for details). In Fig. 1(d) (left panels) we calculate the band structures of TDBG for a twist angle of 1.18° for four different values of electric fields based on this e-h asymmetric model. The corresponding density of states is plotted in the right panels of Fig. 1(d), which depicts various gaps in agreement with those seen from the resistance variation in Fig. 1(b).

We now probe the electron-hole asymmetry in detail by extracting the values of different gaps with varying electric field. Figures 2(a)–2(c) show the variation of resistance as a function of temperature for different electric fields at $n = 0$, $n = -n_s$, and $n = n_s$, respectively. We extract gaps Δ by fitting $R \propto \exp(\Delta/2k_B T)$ at the regime where the resistance decreases exponentially with the temperature. See Sec. III in the Supplemental Material [33] for details of the fits. The R vs T curves show deviation from the Arrhenius nature at low temperature. In particular, we see a shoulderlike variation, which is most prominent for $n = 0$ with D/ϵ_0 stronger than -0.36 V/nm (see Sec. IV in the Supplemental Material [33] for details). In Fig. 2(d) we plot extracted gaps as a function of the perpendicular electric field. Consistent with the band picture we presented in the earlier section, the gap at the charge neutrality point increases with the electric field, while the gaps for $n = \pm n_s$ decrease. An interesting observation is that the gap at CNP starts opening only after a finite electric displacement field of $D/\epsilon_0 \sim 0.2$ V/nm, in contrast to bilayer graphene where the gap opens up as soon as any nonzero electric field is applied (see Fig. S9 in the Supplemental Material [33]). A similar analysis for the device with a twist angle 2.05° shows nonmonotonic behavior of gap with the electric field as the CNP gap closes and reopens with the increasing electric field (see Fig. S8 in the Supplemental Material [33]). In

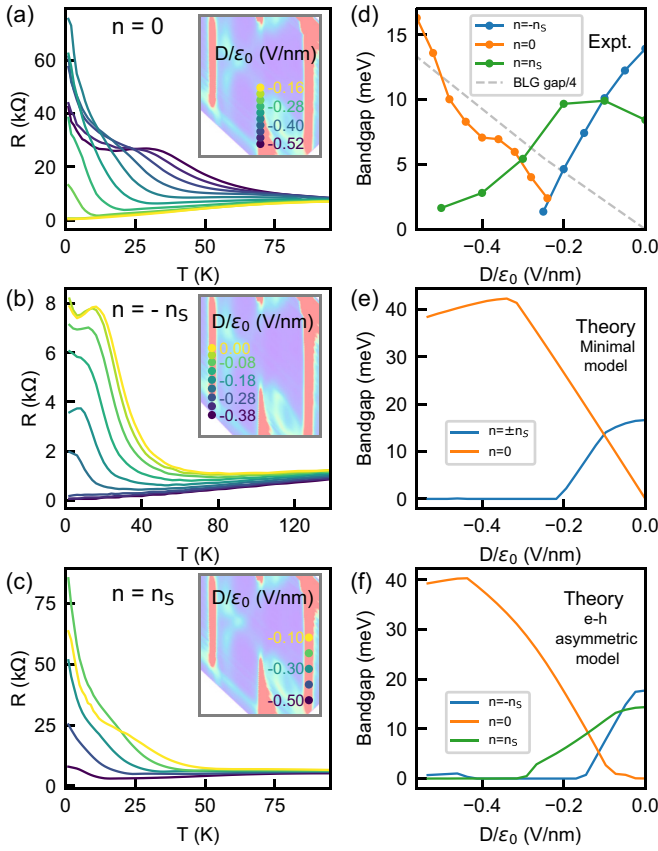


FIG. 2. Temperature variation of resistance at various insulating gaps and estimation of Arrhenius gap. (a)–(c) Line slices of resistance vs temperature for different electric fields at $n = 0$, $n = -n_s$, and $n = n_s$, respectively. Insets show the locations of the data points in (n, D) space. (d) Variation of band gap extracted by Arrhenius fittings to the temperature variation of resistance for $n = 0$ and $n = \pm n_s$. The gap of bilayer graphene, as in Zhang *et al.* [17], is plotted with a dashed gray line to show the dissimilarity between CNP gap in BLG and TDBG. (e) and (f) Variation of the band gap calculated using minimal model (e) and electron-hole asymmetric model (f), respectively; gap opening at CNP only after a critical field seen in experiments is captured only by the electron-hole asymmetric model.

Figs. 2(e) and 2(f) we present the calculated gaps based on the minimal model and the e-h asymmetric model, respectively. The opening of the CNP gap only after $D/\epsilon_0 \sim 0.2$ V/nm shows the inadequacy of the minimal model, which indicates a finite gap at any nonzero electric field. However, the e-h asymmetric model captures the correct trend of the variation of gaps with the electric field for all three different values of $n = 0, \pm n_s$, though overestimating the gaps quantitatively.

In Fig. 3(a) we plot a typical resistance vs temperature curve at $n = 0$ for $D/\epsilon_0 = -0.28$ V/nm when the Fermi energy lies in the CNP gap between two flat bands separated from higher band by moiré gaps as shown in the inset of Fig. 3(a). Clearly there are three distinct temperature regions governed by three different energy scales—the CNP gap, the flat bandwidth, and the separation of higher-energy bands from the Fermi energy. While resistance shows insulating behavior at low temperature (blue) due to activation of carriers

to flat band, at high temperature (red) this is governed by the activation to a higher dispersing band. The thermal activation of charge carriers to higher dispersing bands is also captured by the metal to insulator transition for the states where the chemical potential lies within the flat band, not just at CNP (see Sec. VII in the Supplemental Material [33] for details) [34].

However, there is a mid-range temperature regime (green) where resistance increases with temperature. We now closely examine the temperature regime within the dashed box in Fig. 3(a) and illustrate how the existence of two flat bands separated by a narrow gap can account for this crossover. We employ a simple model, as depicted in the inset of Fig. 3(b), where we consider two narrow bands with bandwidth of w having the constant density of states $\text{DoS}(E) = n_s/w$ with a narrow gap of Δ between them. Here n_s is the number of electrons to fill one such band. We note this model is most appropriately valid before the closure of the moiré gap. As the temperature is increased, the carriers are activated in the narrow bands governed by the Fermi-Dirac distribution, which results in the usual decrease in resistance. However, due to the small energy scales set by w and Δ , the number of activated carriers quickly saturates to the value one would get at a very high temperature, $k_B T \gg \Delta, w$ (see Sec. VI in the Supplemental Material [33]). On the other hand, the mobility keeps on decreasing with increasing temperature and takes over the increase in activated carrier density to show a metal-like increase in resistance.

To quantitatively understand this insulator to metal transition seen in Fig. 3(b), we assume a phonon-scattering dominated temperature variation of the mobility of the form $\mu = a \times T^{-3/2}$. We calculate the conductance using the relation $\sigma = ne\mu$. Here e is the charge of electron and n is the number of activated carriers given by $n = \int \text{DoS}(E) f(E, E_F, T) dE$ with $f(E, E_F, T)$ being the Fermi-Dirac function. We then fit the resultant expression of the conductivity with three fitting parameters, namely, the bandwidth (w), the band gap (Δ), and the overall factor a , to the experimental data. Few such fitting curves are overlaid with the experimental data in Fig. 3(b). The fitting curves deviates at high temperature due to carrier activation to the higher band. The low temperature deviation can be understood since the temperature dependence of the mobility can deviate from $T^{-3/2}$ due to reasons such as disorder scattering at low temperature.

In Fig. 3(c) we plot the extracted bandwidths as a function of the electric field. The bandwidth has a clear trend of increasing with the electric field. This reasonably agrees with the results discussed in Chebrolov *et al.* [19]. The bandwidth has a typical value ~ 1 – 15 meV for mid-range of the electric field where the halo features appear on the electron side. The value of the ratio between electron-electron interaction strength and the bandwidth U/w decreases with electric field from 30 to 2. Here we estimate the Coulomb interaction $U \sim e^2/(4\pi\epsilon_r\epsilon_0\lambda) \sim 30$ meV, ignoring the screening effect and using $\epsilon_r = 4$ [19]. λ is the moiré wavelength for the twist angle 1.18° given by $\lambda = l/[2 \sin(\theta/2)] = 11.9$ nm, with $l = 0.246$ nm being the lattice constant of graphene.

The origin of the electric field tunability of the bandwidth is the subject of detailed theoretical and numerical calculations [19]. However, a simple picture is depicted in the inset of

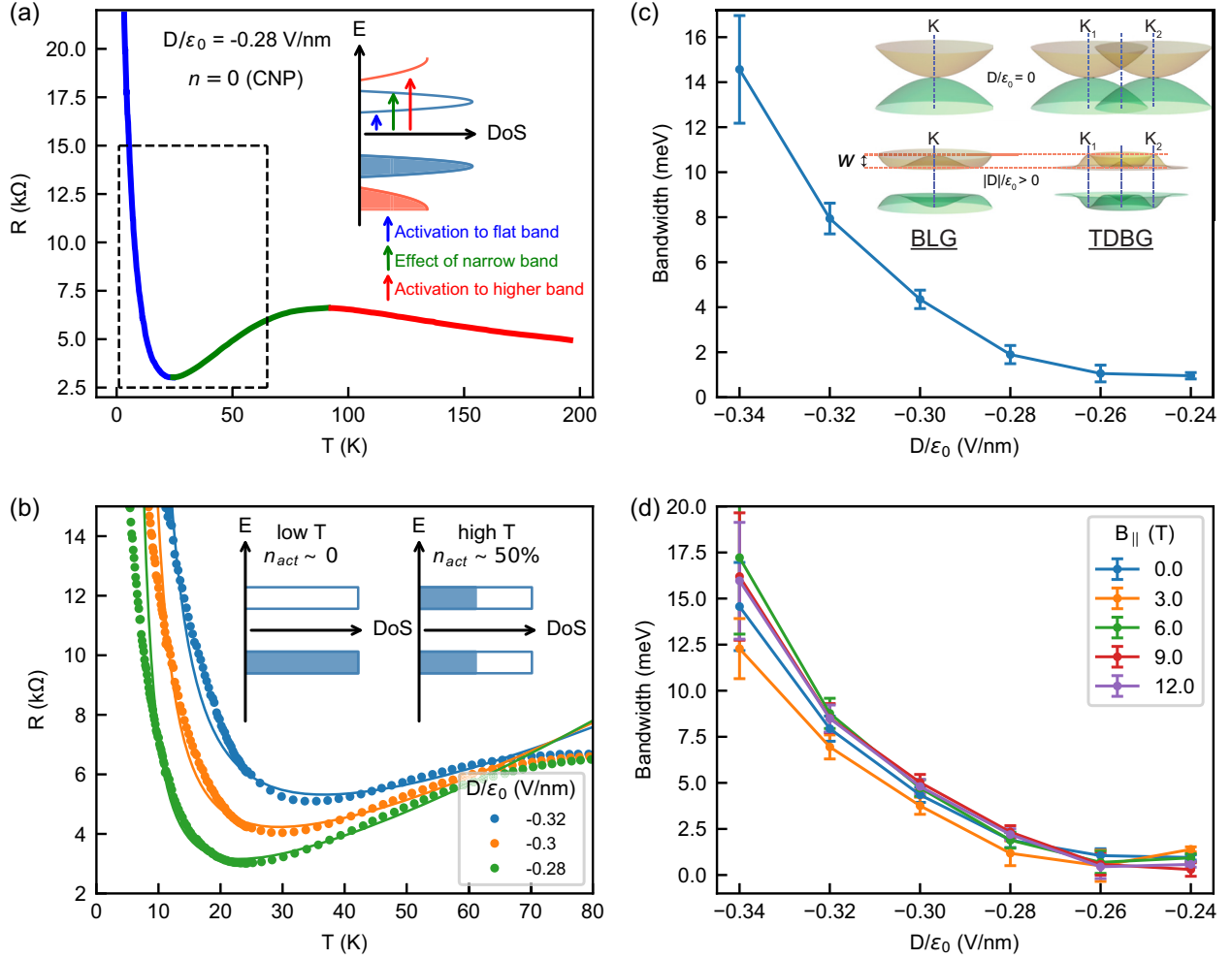


FIG. 3. Insulator to metal crossover—extracting bandwidth. (a) A representative R vs T curve for $n = 0$ depicting three regions of different temperature dependence—insulating behavior at low T (blue), metallic increase (green), and insulating behavior again (red). Inset depicts the three corresponding energy scales in the simple schematic of the underlying band structure with CNP gap and two moiré gaps. The dashed black rectangle indicates the portion of the curve used in (b). (b) Fitting insulator to metal crossover in R vs T curves for different electric fields at $n = 0$ to extract bandwidth. Points correspond to measured resistance and the solid lines are the fits. Inset shows the simple model of two flat bands with a small gap used in the extraction. The number of activated charge carriers saturates to 50% at higher temperature from its low T value of $n_{act} \sim 0$. (c) The variation of extracted bandwidth (w) with the electric field at $B_{||} = 0$. The inset shows how TDBG can inherit the tunability of its bandwidth from BLG. (d) The variation of bandwidth (w) with the electric field for different in-plane magnetic fields.

Fig. 3(c) considering the tunability of underlying BLG band structure. Upon applying the electric field, the low energy part of the quadratic bands in BLG forms a “Mexican hat” structure with local maxima and minima around K points in the Brillouin zone [15,18]. The depth of the Mexican hat scales approximately as the cube of the electric field. A small angle twist of two copies of BLG in TDBG effectively hybridizes such adjacent $E - k$ cones and the bandwidth of the low energy flat band inherits the electric field dependence of the depth of the Mexican hat in BLG. The tunability of this energy scale may be enhanced in TDBG from that of BLG due to additional hopping terms and modified effective mass.

Finally, we study the effect of in-plane magnetic field on the bandwidth, motivated by its profound role to enhance correlated gaps as already seen in Fig. 1(c). In Fig. 3(d) we plot the variation of bandwidth with electric field for different magnitude of in-plane magnetic field. The overlapping

error bars suggest there is no clear trend implying that the bandwidth is not substantially tuned with in-plane magnetic field.

III. CONCLUSIONS

We find TDBG to be a highly tunable system with moiré gaps that can be closed with increasing electric field while the flat bands split into two; eventually, the two narrow flat bands merge with the higher dispersing bands. A plethora of novel phenomena have been identified to be originating from the underlying flat bands in the twisted graphene system, without directly quantifying the flatness of the bands using transport. In strongly correlated systems, the bandwidths and gaps can be modified because of interactions. Our measurement and subsequent analysis provide a route to quantitatively

establish tunability of bandwidth with electric field in flat band systems.

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