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RAPID COMMUNICATION

Observation of quadratic magnetoresistance in twisted double bilayer graphene

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Magnetoresistance (MR) provides rich information about Fermi surface, carrier scatterings, and exotic phases for a given electronic system. Here, we report a study of the magnetoresistance for the metallic states in twisted double bilayer graphene (TDBG). We observe quadratic magnetoresistance in both Moiré valence band (VB) and Moiré conduction band (CB). The scaling analysis shows validity of Kohler's rule in the Moiré valence band. On the other hand, the quadratic magnetoresistance appears near the halo structure in the Moiré conduction band, and it violates Kohler's rule, demonstrating the MR scaling related to band structure in TDBG. We also propose an alternative scaling near the halo structure. Further analysis implies that the observed quadratic magnetoresistance and alternative scaling in conduction band are related to the halo boundary. Our results may inspire investigation on MR in twisted 2D materials and provide new knowledge for MR study in condensed matter physics.

Keywords: twisted double bilayer graphene, Kohler's rule, magnetoresistance scaling, critical behavior

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Magnetoresistance (MR) is a measure of resistance to the change of magnetic field, and it provides rich information about Fermi surface, carrier scatterings, and exotic phases for a given electronic system.[1–4] MR is quantified as MR $= [R_{xx}(B) - R_0]/R_0$, where R_0 is the resistance at zero magnetic field. From Drude formula, the MR in metals is usually quadratic in magnetic field B. More generally, it follows the Kohler's rule.[1,3,5] that MR in metals scales as a function of the ratio B/R_0, i.e., MR $= f(B/R_0)$. Kohler's rule has been found to be valid in various materials including metals, semimetals, and cuprate superconductors.[6–11] Meanwhile, the violation of Kohler's rule has also been widely observed and often related to Fermi surface reconstruction, magnetic breakdown, anisotropic effects, quantum correction to resistance, and other carrier scattering mechanisms (phonon scattering or electron–electron interaction),[1,3,12–16] reflecting emergent physics. One example is the strange metal phase observed in iron pnictide superconductor, showing MR scalable by energy scale.[17] Thus, the violation of Kohler's rule and the emergence of new scaling usually signals underlying exotic physics, deserving in-depth investigation.

Recent years, twisted Moiré systems have been demonstrated as an ideal platform hosting lots of unconventional states, such as correlated insulators,[18–24] superconductivity,[19,25,26] ferromagnetism,[27–30] Pomeranchuk effect,[31,32] quantum criticality.[33–35] However, most works in these Moiré systems focus on emergent ground states in the strong correlated regime,[36] leaving the metallic states and its MR poorly explored.

In this article, we report a study of magnetoresistance behaviors in metal state of twisted double bilayer graphene (TDBG). In TDBG, the Moiré conduction band (CB) in TDBG is much narrower than the Moiré valence band (VB), with the former holding correlated states but the latter not.[20–22,37] In addition, the displacement field D could *in situ* tune Moiré band bandwidth. Thus, TDBG is a good platform to design contrast experiments in one device to avoid sample variations. We observe quadratic magnetoresistance in both CB and VB. Kohler's rule is found to be valid in the valence band. By contrast, in the conduction band, we observe a violation of Kohler's rule and propose an alternative scaling rule near the halo structure. We find a close connection between the violation of Kohler's rule and the halo boundary.

The twisted double bilayer graphene samples were pre-

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pared by "cut and stack" technique[38] and fabricated into Hall bar geometry.[35] "Mechanical squeeze" technique is introduced to remove nanobubbles and contaminations between layers.[19,39] The dual gate geometry enables us to tune carrier density n and electric displacement field D independently via carrier density $n = (C_{BG}V_{BG} + C_{TG}V_{TG})/e$ and displacement field $D = (C_{TG}V_{TG} - C_{BG}V_{BG})/2\varepsilon_0$, where C_{BG} and C_{TG} are the geometrical capacitance per area for bottom and top dielectric layers, V_{BG} and V_{TG} are the bottom and top gate voltages, e is the elementary charge, and ε_0 is the vacuum permittivity. Here, $n_s \approx 8\theta^2/\sqrt{3}a^2$ is the carrier density at full filling of the first Moiré band and the Moiré filling factor $\nu = 4n/n_s$ is the number of carriers in one Moiré unit cell, with a being the lattice constant of graphene. For the 1.28° TDBG device, the transfer curves measured in different pairs of Hall bar indicate a small twist angle variation ($\Delta\theta \sim 0.03°$) over a span of about 10 μm (Fig. S1). The 1.28° TDBG device holds half-filling correlated insulator states with commensal halo structure in conduction band (Fig. 1(a)), in agreement with the previous studies.[20–22,37] Here we mainly focus on the D dependence of MR, since the carrier density (n) dependence of magnetoresistance (Fig. S2) is rather complex.

Firstly, we focus on magnetoresistance data in the Moiré valence band. Plots of symmetrized longitudinal resistance R_{xx} against B are shown in Fig. 1(b), where R_{xx} increases from ~ 200 Ω to ~ 2000 Ω nonlinearly as the B changes from 0 to 2 T, and the curves with lower zero-field resistance have stronger B response. By transforming R_{xx} into magnetoresistance (MR) through $\mathrm{MR} = [R_{xx}(B) - R_0]/R_0$ and changing the x-axis from B to the square of B, the MR curves show quadratic-in-B dependence at low field, as shown in Fig. 1(c). Figure 1(d) shows the Kohler plot of the curves from Fig. 1(c). The data from different D collapse into one curve, demonstrating the validity of Kohler's rule in VB. The observation indicates normal metal behaviors in VB. However, the validity of Kohler's rule is observed in a limited range of D from ~ 0.58 V/nm to ~ 0.74 V/nm (Fig. S3). We think that it is related to complexity of band structure in TDBG, such as van Hove singularity, crossing of VB with CB at lower D, and valley polarization at higher D.[40]

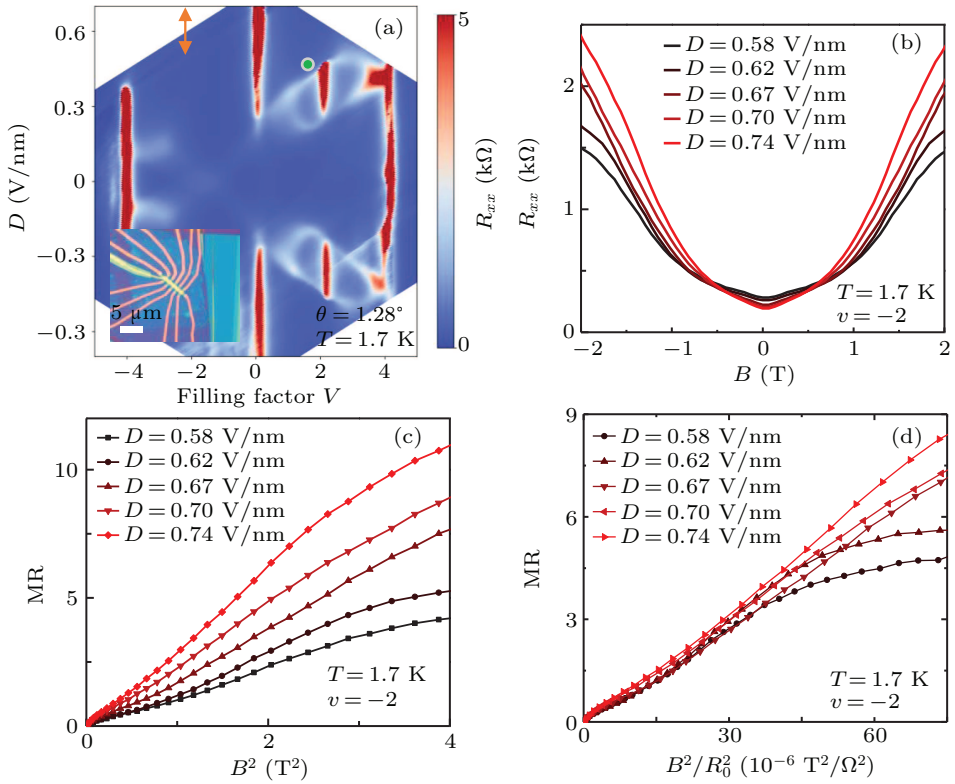


Fig. 1. Validity of Kohler's rule in valence band of 1.28° TDBG. (a) Resistance mapping against filling factor ν and displacement field D. The inset shows the optical microscope image of this device. Scale bar: 5 μm. The green dot and orange two-end arrow line mark the position of MR studied in the main text. (b) Longitudinal resistance R_{xx} against magnetic field at representative D in valence band. (c) MR against the square of B taken from (b). (d) Kohler's plot of the MR curves in (c).

The quadratic magnetoresistance is also observed in CB of the same device. We focus on magnetoresistance outside the halo structure, and MR inside the halo will be discussed in the supplementary information [Figs. S8(d)–S8(f)]. The halo boundaries at $\nu = 1.75$ corresponds to displacement field $D_{C1} = 0.417$ V/nm and $D_{C2} = 0.209$ V/nm. Figure 2(a) shows plots of $R_{xx}(B)$ at representative D that are larger than D_{C1}. The anti-symmetrized Hall resistance R_{xy} at low field is insensitive to the change of displacement field, as shown in Fig. 2(b), ensuring the normal metal state. The R_{xx} increases

nonlinearly with B, and the increase becomes slower at larger D. The data from Fig. 2(a) shows a quadratic MR in Fig. 2(c). By doing Kohler's plot, however, these quadratic curves do not converge to a single plot, implying a violation of Kohler's rule [Fig. 2(d)].

Then we investigate Kohler's rule further and propose an alternative scaling. For quadratic MR, it follows $R_{xx}(B) = A_2 \times B^2 + R_0$. The validity of Kohler's rule requires $\mathrm{MR} = (A_2 \times B^2)/R_0 \propto (B/R_0)^2$, so the B-square coefficient A_2 should be inversely proportional to zero-field resistance R_0, namely $A_2 \propto 1/R_0$,[1] which is in line with the observation in VB (Fig. S4). By extracting coefficients from Fig. 2, A_2 and R_0 are found to be positively linearly correlated in Fig. 3(a), and it leads to the observed violation of Kohler's rule in CB. What's more, if we put the linear dependence $A_2 = C \times R_0 + F$ into $\mathrm{MR} = (A_2 \times B^2)/R_0$, we can reach $\mathrm{MR} = (C + F/R_0)B^2$. This demonstrates an alternative MR scaling (we call it the "alternative scaling" hereafter for simplicity) based on the observed linear dependence between A_2 and R_0. Here we have $C = (0.658 \pm 0.035)\ \mathrm{T}^{-2}$, $F = (-289.69 \pm 18.62)\ \Omega \cdot \mathrm{T}^{-2}$ taken from the data in Fig. 3(a). Applying this scaling to MR curves in Fig. 2(c), Fig. 3(b) shows all MR curves collapse onto a single one trace at low field, demonstrating the validity of the alternative scaling we suggest. Meanwhile, the scaled trace holds slope about unity, consistent with the requirement of $\mathrm{MR} = (C + F/R_0)B^2$. We emphasize that the alternative scaling is found to be valid near the halo structure. At an even larger $D > 0.47$ V/nm (further away from the halo structure), the resistance becomes insensitive to B (ΔR is the order of several tens of Ω, as shown in Fig. S6), making the scaling unavailable. Morever, the normal metal state at another side of the halo structure ($D < D_{C2}$) in the same device shows the validity of alternative scaling (Fig. S5). Similar behavior is observed in another 1.32° TDBG device, both inside and outside the halo in D dependence, confirming the effectiveness of the alternative scaling (Fig. S8).

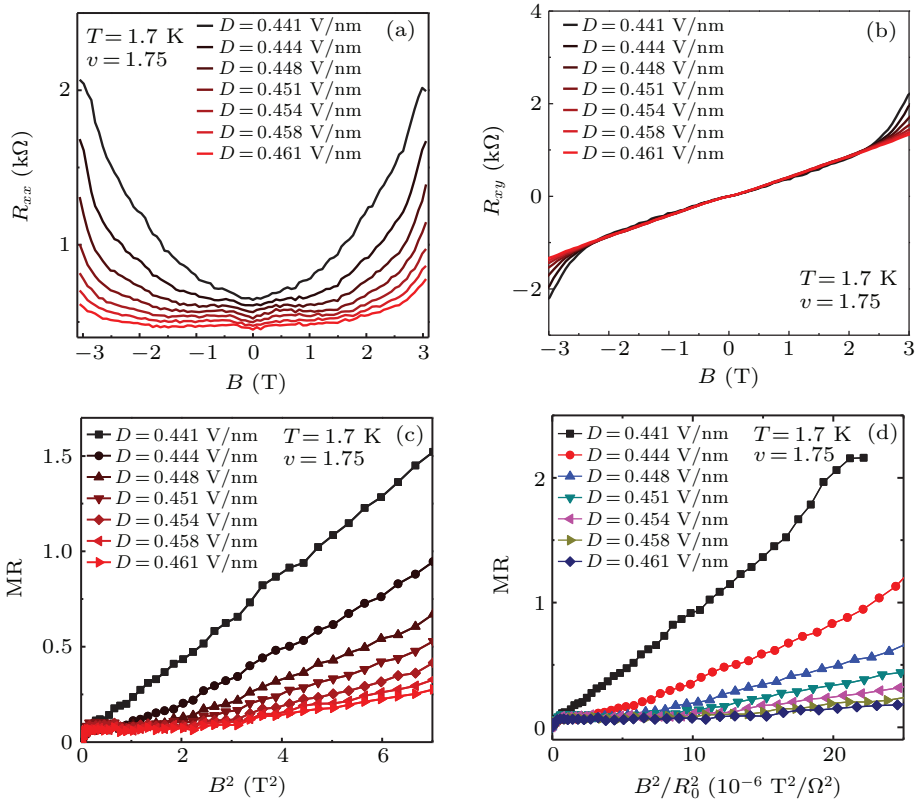


Fig. 2. Violation of Kohler's rule in conduction band of 1.28° TDBG. (a) Longitudinal resistance R_{xx} against magnetic field B at representative displacement fields D, with ε_0 being the vacuum permittivity. (b) Hall resistance R_{xy} against B at representative D corresponding to (a). (c) MR against the square of B taken from (a). (d) Kohler's plot of the MR curves in (b).

To clarify the origin of alternative scaling in CB, we can discuss about the violation of Kohler's rule (Fig. 2(d)) at first. The Boltzmann theory is quasi-classical and based on single-particle assumption, neglecting quantum mechanical effects and many-body effects. The deduction of Kohler's rule is based on the idea of the presence of a universal mean free path.[3] Consequently, the violation of Kohler's rule often results from magnetic breakdown, quantum correction of resistance, anisotropic effects in systems, phonon scattering, and el–el interaction.[1,3,12–16] In this paper, all magnetoresistance data are acquired in the low-field limit ($\mu B < 1$, and no sign of magneto-oscillation is observed), so magnetic breakdown can be safely ruled out. Due to the validity of Kohler's rule in valence band in the same device, anisotropic effects such as mag-

netic impurity, strain[41] cannot be the answer. Quantum correction of resistance means weak localization effect or weak anti-localization effect, while no obvious such effect is observed near the halo structure (Fig. 2(a)), thus the contribution of quantum correction should be limited. At our base temperature ($T \sim 1.7$ K), the electron-phonon scattering should not dominate due to the low occupation number of phonons.[42] The violation of Kohler's rule and the validity of alternative scaling in conduction band may root in el–el interaction near the halo structure.

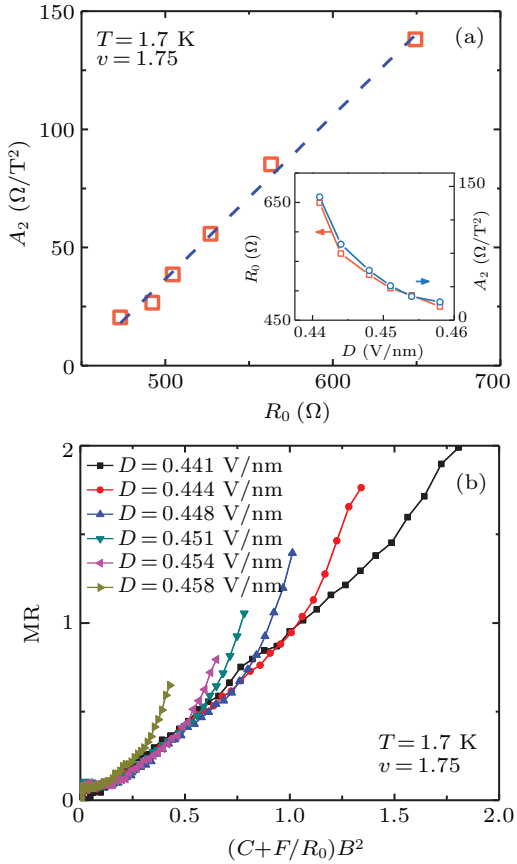


Fig. 3. The alternative scaling of magnetoresistance in TDBG. (a) B-square coefficient A_2 against zero-field resistance R_0 taken from Fig. 2(a). The blue dashed line shows linear fit between A_2 and R_0. Inset: A_2 and R_0 against D used in the main figure. (b) The alternative scaling of MR curves in Fig. 2(c) with linear fitting $A_2 = C \times R_0 + F$. The slope of the scaled trace is about unity, in agreement with the requirement of MR $= (C + F/R_0)B^2$ in the main text.

We also find that the quadratic magnetoresistance and alternative scaling in CB are related to the halo boundaries. In our previous work, we found the T-square coefficient of resistance diverges at the halo boundaries (Fig. S7), which signals the enhancement of effective mass at the halo boundaries.[35] Here we demonstrate that the B-square coefficient A_2 also tends to diverge near the halo boundary where the alternative scaling works [inset of Fig. 3(a), and Fig. 4]. The halo boundary acts as phase boundary of symmetry broken metal state inside it,[37] so we think the quadratic magnetoresistance and alternative scaling root in the critical behavior at the halo boundaries. The specific reason may be el–el interaction enhanced by maximized effective mass there.[35] Recently, an STM study on TDBG reports interaction-driven electronic nematic phase in the normal metal state,[43] in agreement with our hypothesis about the effect of el–el interaction in normal metal state of TDBG. Taking the above discussions together, we suggest that the halo boundaries are relevant to the alternative scaling and the corresponding quadratic magnetoresistance in Moiré conduction band of TDBG. This is beyond classical Boltzmann theory and calls for future theoretical studies on it.

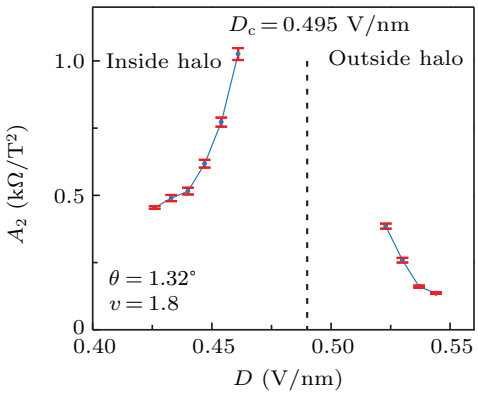


Fig. 4. The diverging A_2 near the halo boundary. The extracted A_2 against D near the halo boundary in the 1.32° TDBG device. The A_2 tends to diverge as approaching the halo boundary both inside and outside the halo, showing the correlation between the halo boundary. The A_2 is taken from MR curves in Fig. S8.

In conclusion, we have studied the magnetoresistance in the metal state of TDBG. The longitudinal resistance shows quadratic-in-B dependence in the low-field limit in both Moiré conduction band and valence band. Meanwhile, the quadratic magnetoresistance in Moiré valence band follows Kohler's rule, consistent with requirement of classical normal metal. However, we find a violation of Kohler's rule in conduction band near the halo structure and propose an alternative scaling based on the positive linear dependence between A_2 and R_0. Combining the validity of Kohler's rule in valence band, the quadratic-in-T resistance and diverging A_2 near the halo boundaries, we find that the quadratic magnetoresistance and alternative scaling in the conduction band are related to the halo boundaries. The enhanced effective mass near the halo boundaries may be responsible for this finding. Our paper will provide a new perspective to study twisted 2D material systems and quadratic magnetoresistance in condensed matter physics.

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