

Supplementary information: Breakdown of semiclassical description of thermoelectricity in near-magic angle twisted bilayer graphene

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I. Device fabrication

In this study, we have fabricated the devices in two different configurations. In the first configuration (denoted by H1) two graphene layers are aligned at an angle θ and encapsulated within two hexagonal boron nitride (hBN) (Fig. S1(a)). The device is etched into a hall bar and consists of both single layer graphene (SLG) and twisted bilayer graphene (tBLG) regions. A local gold top gate is fabricated only on the tBLG region, while the global silicon backgate is used to tune the number density (n) in SLG region. The device schematic and optical micrograph in this geometry are shown in Fig. S1(b), (c). In the second configuration (denoted by C1), the tBLG device consists of two graphene layers aligned at $60^\circ + \theta$, thus θ being the effective twist angle [1], and encapsulated within two sheets of hBN (see schematic shown in Fig. S1(d)). The moiré super-lattice is formed at the overlap region ($\approx 5 \mu\text{m} \times 6 \mu\text{m}$), and the monolayer branches of graphene on four sides act as electrical leads. The device micrograph is shown in Fig. S1(e). A local top-gate tunes n of the overlap region. Fig. S2 gives the details of all the different devices used in this study.

We have used Raman spectroscopy to qualitatively differentiate between large and small twist angle tBLG devices and to avoid making devices on tBLG stacks which turn into bilayer graphene (BLG) after the transfer process. Specifically, the shape and position of the $2D$ peak is sensitive to the twist angle and provides a rough estimation[2–4]. Fig. S1(f) shows the Raman spectra of $\sim 1.6^\circ$, $\sim 2.4^\circ$, $\approx 4^\circ$ tBLG devices and single layer graphene (SLG). $2D$ peaks of small twist angle devices have higher values of the full width half maximum (FWHM) than SLG and usually have an additional shoulder. In contrast, large twist angle tBLG has a spectrum very similar to SLG as the two layers are decoupled. Though the Raman spectrum does not have enough resolution to precisely determine the twist angle, it provides a qualitative information about the angle. We have used transport methods to calculate the exact twist angle for all devices except D5 ($\theta \sim 4^\circ$), where Raman spectrum is used to estimate the approximate angle.

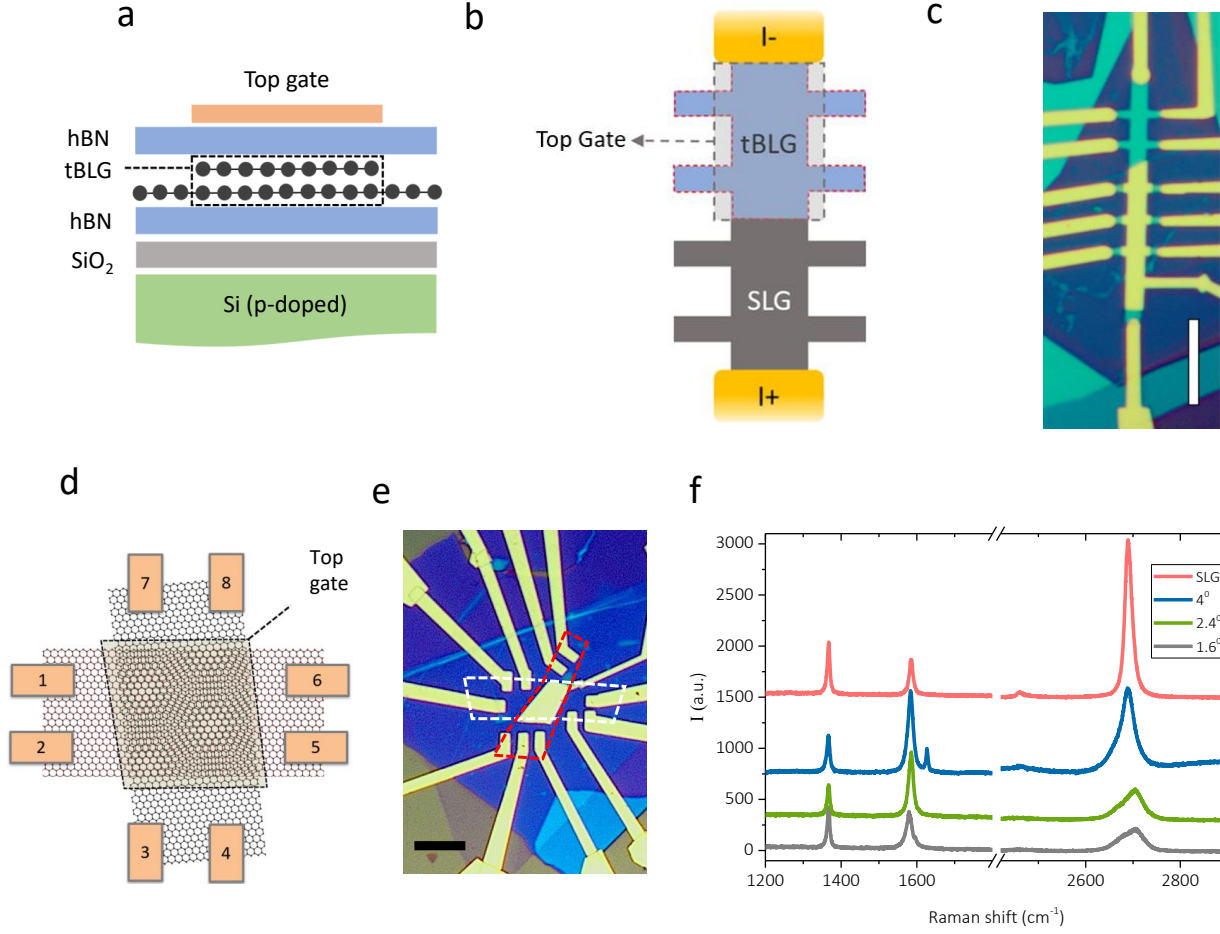


FIG. S1: (a) Schematic of the cross-sectional view of the device showing the constituent layers. Devices were fabricated in two different geometries, denoted by H1 and C1. (b) Schematic of the device fabricated in hall bar geometry (H1). The dashed rectangle shows the tBLG region covered with gold top gate while the gray region represents the SLG region. (c) Optical micrograph of a typical tBLG device fabricated in H1 geometry. The scale bar represents a length of 5 μm . (d) Device schematic representing the cross-bar geometry (C1). The dashed region shows the tBLG region covered with top gate (e) Optical micrograph of a tBLG device fabricated in G1 geometry. The scale bar represents a length of 5 μm and dotted lines show the graphene edges. (f) Comparison of Raman spectra for G peak and 2D peak obtained for $\approx 1.6^\circ$, $\sim 2.4^\circ$, $\approx 4^\circ$ and single layer graphene with relative offset in the intensity for clarity.

Experimental details of the different devices in this study

Device	Twist angle ($^{\circ}$)	Geometry
D1	1.01	H1
D2	1.24	H1
D3	1.6	C1
D4	1.7	H1
D5	~ 4	C1

FIG. S2: Twist angle and device geometry of different samples measured in this study.

II. Quantum oscillations in D2 and D3 devices

Fig. S3(a) shows the resistance (R) measured at 5 K in the absence of an external magnetic field, and Fig. S3(b) shows the results of quantum oscillations measurements in 1.6° device. We have plotted the first derivative of R with respect to the gate voltage tuned from Dirac point ($V_{tg} - V_D$) in order to get a better colour contrast. The quantum oscillations emerging from the CNP are eight fold degenerate as previously reported for devices with similar twist angles[5]. In contrast, the Landau fans emerging from $\nu = \pm 4$ are four fold degenerate, accounting for spin and Fermi-contour degeneracy. Additionally, we have estimated the twist angle using the quantum oscillations, which comes out to be $\approx 1.6^{\circ}$

Fig. S4(a) shows the derivative of the resistance (dR/dV_g) versus gate voltage and $B_{\perp}$ in device D2. We plot the Landau level structure deduced from the oscillations in Fig. S4(b). At low field, The black Landau fan, which originates from the charge neutrality point (CNP), and the pink Landau fan, which originates from the superlattice density, show the filling-factor sequences $\pm 4, \pm 8 \dots$ as expected for single-particle band structure with four-fold spin and valley degeneracies. We note that as the magnetic field is increased above 5 T, either valley or spin degeneracy breaks and two faint lines with filling-factor ± 2 appear to emerge from the CNP. Additionally, we find multiple other Landau fans (purple lines) emerging from intermediate densities and are not expected from single-particle band structure. The origin of these lines can be traced back to the resistive peaks observed near integer fillings as shown in Fig. S4(c)

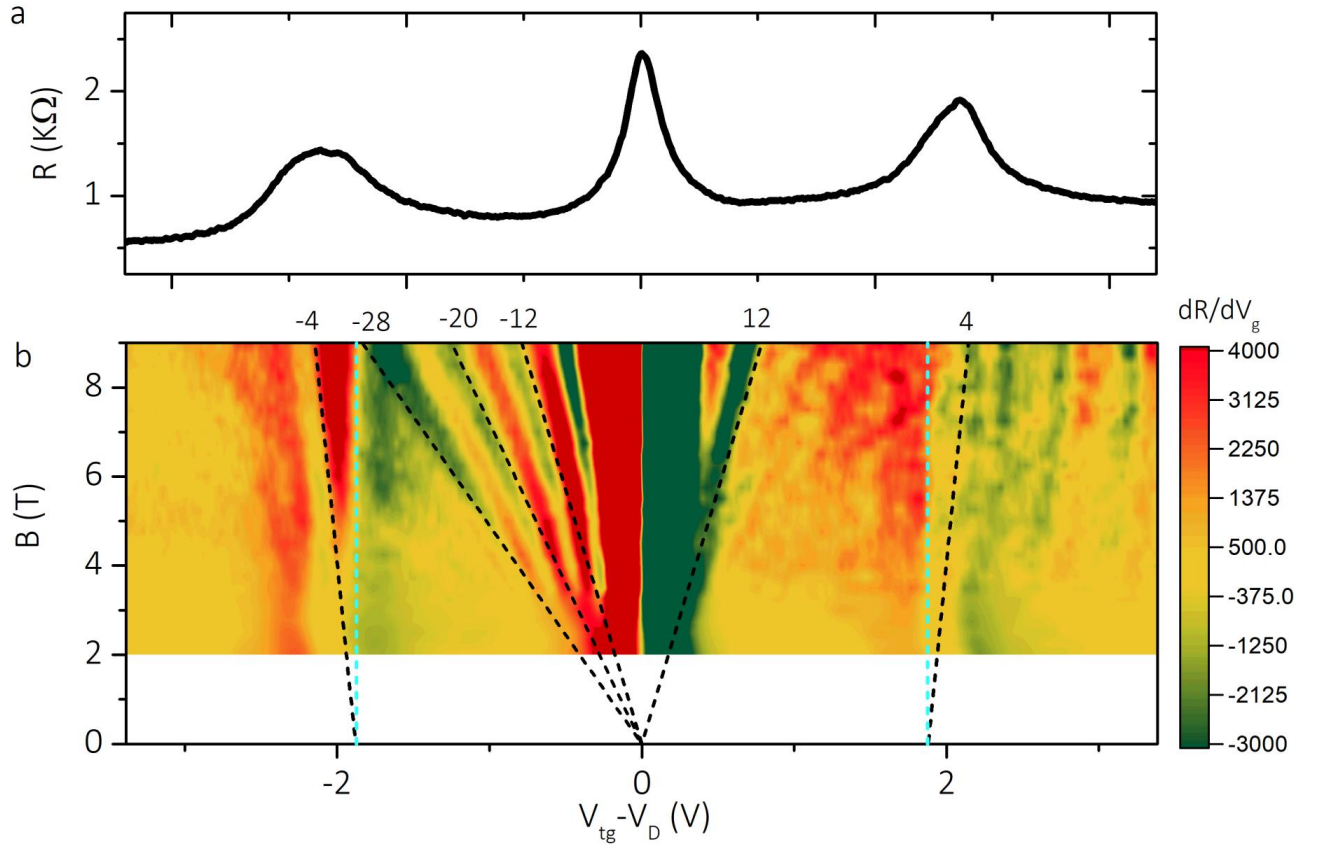


FIG. S3: (a) R as a function of n measured at 5 K. (b) Quantum oscillations in 1.6° device. The first derivative of R with respect to gate voltage is plotted in order to enhance the colour contrast. Black dashed lines show the Landau fans emerging from the CNP and $\nu = \pm 4$.

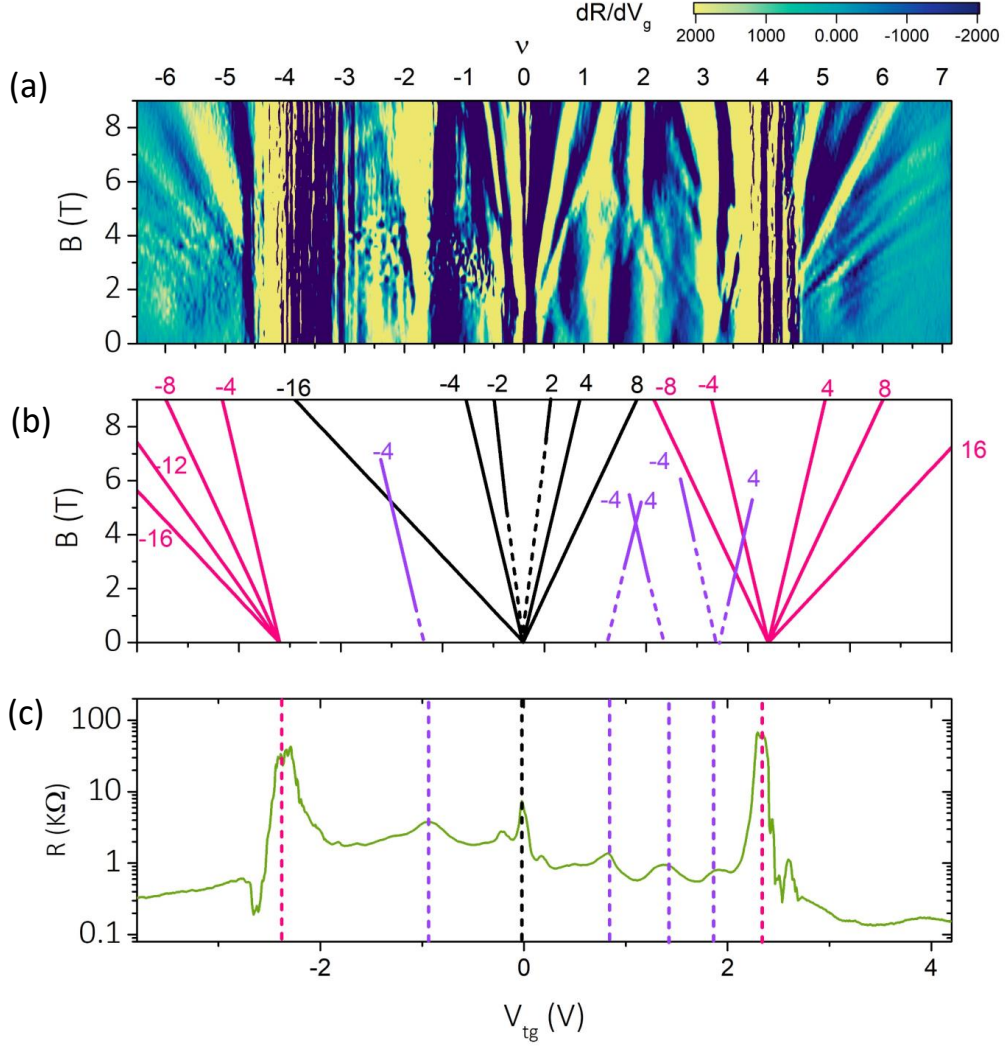


FIG. S4: (a) Quantum oscillations in device D2 (1.24°) measured at 100 mK. (b) Black and pink solid lines show the Landau fans emerging from the CNP and $\nu = \pm 4$, whereas purple lines show fans originating from intermediate fillings, where peaks in R are observed. (c) R as a function of V_{tg} at zero magnetic field. Dashed lines correspond to resistive peaks which give rise to Landau fans.

III. Additional data on electrical transport in different devices

A. Device geometry and transport measurements in device D1

Device D1 was fabricated with a multilayer WSe_2 between the top hBN and the tBLG (Fig. S5 (a)), and the optical micrograph of the device is shown in Fig. S5(b). We observe weak resistive features at $\nu = 1, 2$ and 3 at 5 K, which persist upto 20 – 25 K (denoted by CS) and show a clear metallic behaviour (Fig. 1a). Additionally resistance peak at $\nu = 9$ (Fig. S5 (c)) indicates the presence of correlations even in higher bands which have not been studied extensively so far and requires further investigation.

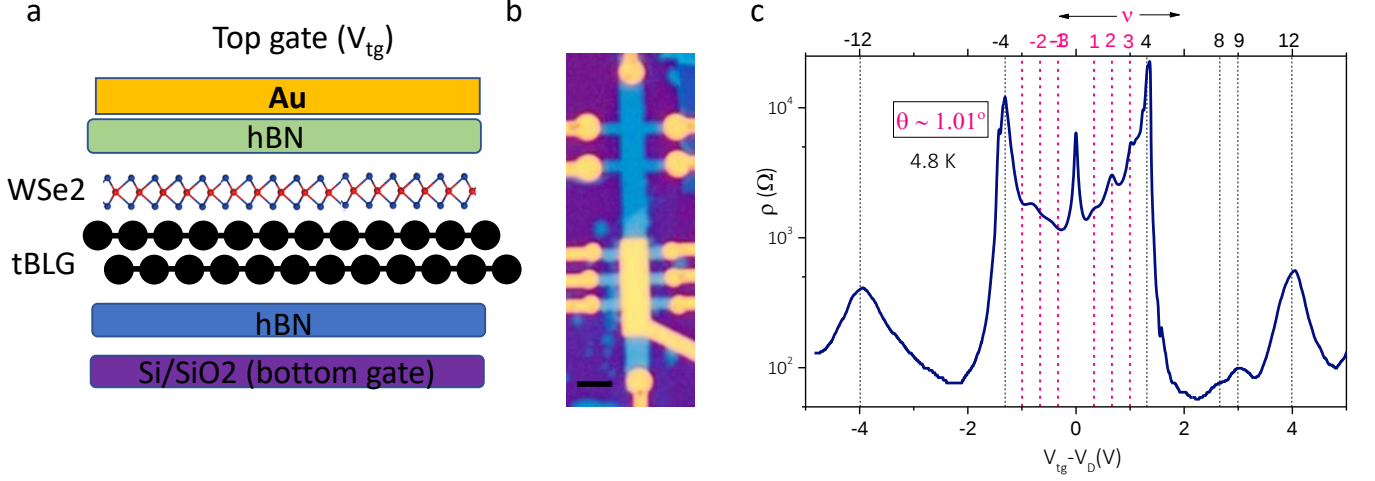


FIG. S5: (a) Schematic cross-sectional view of the device consisting of a multilayer WSe₂ sandwiched between the top hBN and the tBLG. (b) Optical micrograph of the device. The scale bar represents a length of 2 μm . (c) ρ as a function of gate voltage measured at 4.8 K.

B. Temperature dependence of resistance in device D2

Fig. S6(a) shows the resistivity ρ measured in the 1.24° device as a function of n and T . We observe three resistive peaks near integer fillings on the e-side. The peaks near $\nu = 1, 2$ show a weak insulating behaviour upto 30 – 35 K (Fig. S6(c)), whereas the peak close to $\nu = 3$ show a metallic dependence throughout the temperature range. Similarly on the h-side, we observe a resistive peak between $\nu = -1$ and -2 , which shows an insulating behaviour upto 40 – 50 K. These peaks can not be explained in single particle band picture and are attributed to e-e correlations present in the system. In the following discussion, we denote the peaks near $\nu = 1, \pm 2$ as CI and the metallic state near $\nu = 3$ as CS. To further understand the origin of these CI/CS features, we focus on $\rho - T$ data between 0 and 3 electrons per superlattice in Fig. S6 (b). We notice that the peak near $\nu = 1$ starts moving gradually and at $T = 30 - 35$ K, it moves to $\nu = 1$. Similar shift of resistive peak can be traced near $\nu = 3$, however the peak near $\nu = 2$ does not show any substantial change in its position. This observed effect has previously been reported for magic tBLG angle devices and is attributed to isospin Pomeranchuk effect arising due to small isospin stiffness in these systems [6, 7], however the origin of such shifts in ρ peaks is still not very clear and requires further understanding.

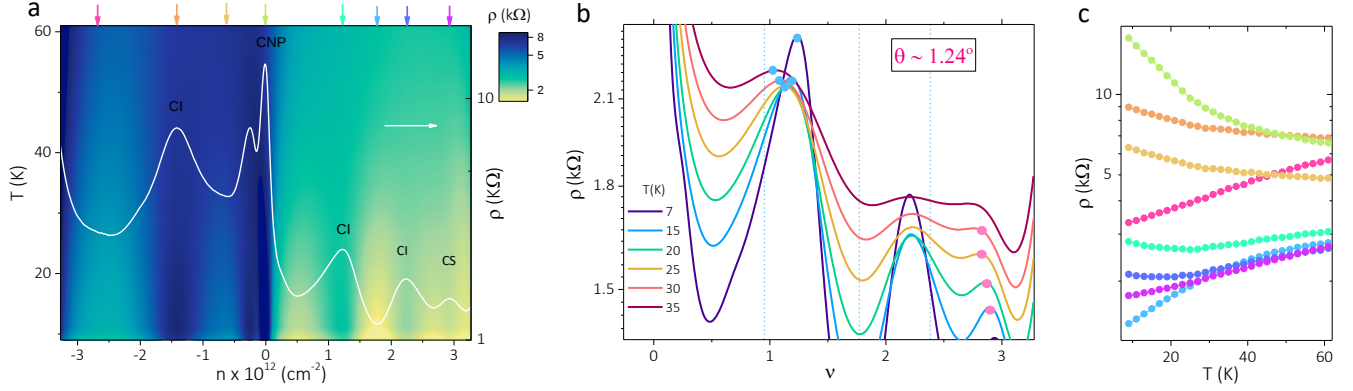


FIG. S6: (a) Temperature dependence of ρ in the 1.24° device, which exhibits multiple correlated resistive features close to integer fillings. The Right axis shows the R measured at 9 K. (b) ρ as a function of carrier density at a few selected temperatures. Light blue and pink circles indicate the shift in peak positions at $\nu = 1$ and 3. (c) $\rho(T)$ measured at different densities as marked by the arrows at the top in (a).

C. Resistance measured in different configurations for device D3 (1.6°)

We have measured the electrical resistance in multiple contact configurations to probe the twist angle inhomogeneity in our device. Optical micrograph of the device along with the contact configuration is shown in Fig. S7(a), where the dashed lines show the two graphene layers and the metal top gate. Fig. S7(b) shows a comparison of four terminal resistance measured at 3 K in three different configurations. Even though the magnitude of the resistance is slightly different in various configurations, the device exhibits a consistent moiré unit size in all configurations, as is evident by the good alignment of insulating states ($\nu = \pm 4$), thereby ensuring angle homogeneity. Further, we have measured the temperature dependence of resistance in the configuration given by, $I : 3 - 8, V : 2 - 9$. Fig. S7(c),(d) show the resistance in the T linear regime. We find that R is metallic at $T \lesssim T_H$, where $T_H \sim 100 - 200$ K is a doping-dependent characteristic temperature. The results obtained in this configuration are consistent with the transport data shown in Fig. 1. Furthermore, it is to be noted that results shown in Fig. S7(c),(d) are from a different thermal cycle as compared to Fig. S7(b).

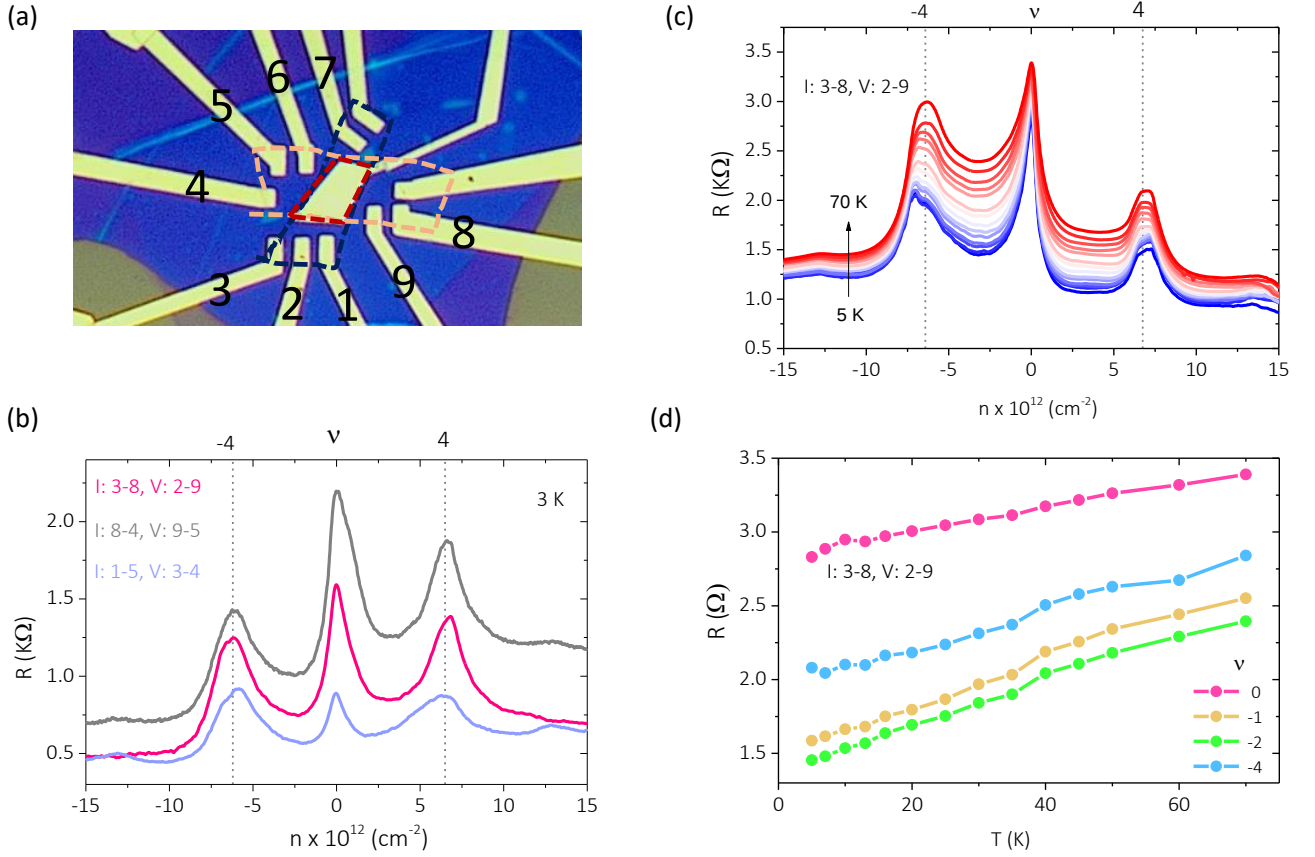


FIG. S7: (a) Optical micrograph of the device D3 with the contact configuration. (b) Measured resistance (R) for three different configurations. Dashed lines show filling factor $\nu = \pm 4$. (c) R as a function of density at different T ranging from 5 K to 70 K in the configuration marked by, $I : 3 - 8, V : 2 - 9$. (d) T -dependence of R at different filling factors (ν) in the T -linear regime.

D. Electrical transport in different thermal cycles in device D3

Fig. S8(a) shows the transport data from the 1.6° device in one of the thermal cycles (labelled as ‘a’), where the resistance shows a shallow peak near $\nu = \pm 2$. We observe a clear electron-hole asymmetry in the R peaks, which is likely to emerge from the asymmetric band structure of tBLG. Similar shallow peaks in resistance reported in previous studies are attributed to van Hove singularities in the non-interacting DOS which in-turn lead to large scattering cross-section for quasiparticles [8, 9]. However, we notice that the R peaks are suppressed by the application of a parallel magnetic field and finally vanishes at a field of $6(2) \text{ Tesla}$ for $\nu = 2(-2)$ (Fig. S8(b)), which is similar to the effects seen in previous studies on magic angle tBLG [10]. Furthermore, we have measured the T -dependence in a small range to probe the origin of these peaks (Fig. S8(c)). Although the magnitude of the peak decreases at higher T ($7 - 8 \text{ K}$), it is not possible to extract any quantitative information from the T dependence because of limited data. However, these features almost vanish after a thermal cycle to the sample (labelled by ‘b’) as shown in Fig. S8(d). Observation of such features in R along with a clear violation of Mott formula indicate the role of interactions. However the origin of these peaks in R is still not very clear at this point.

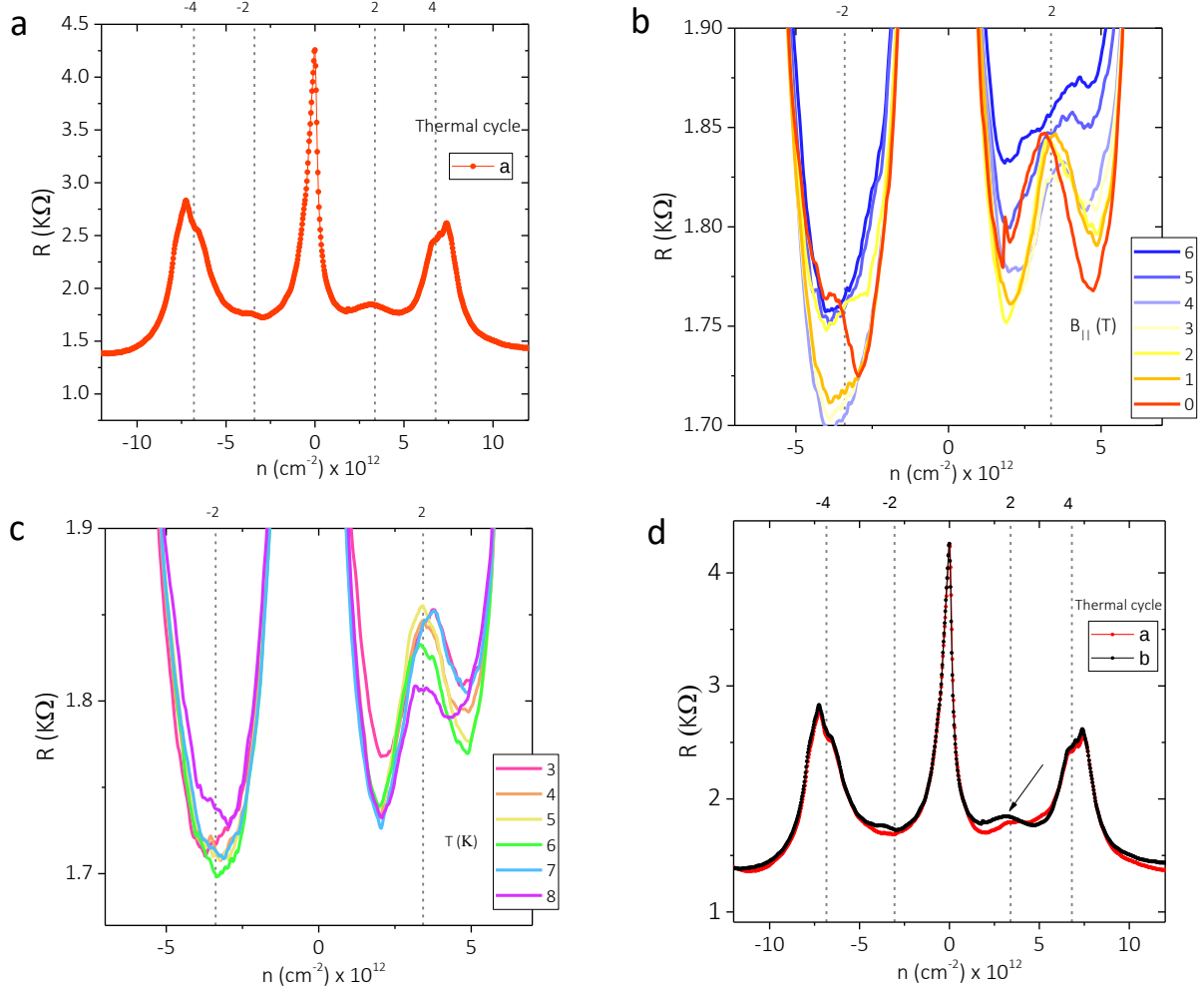


FIG. S8: . (a) R as a function of n for device D3 in thermal cycle number 'a' (b) Parallel Magnetic field dependence of R , zoomed-in to focus on R peak at $\nu = \pm 2$. (c) T dependence of resistance peak near $\nu = \pm 2$ (d) R plotted for two different thermal cycles.

E. Temperature dependence of resistance in device D3

Fig. S9(a),(b) show ρ in the T -linear regime across a wide range of carrier density between the CNP and $\mp n_s$. In this T range, transport can be assumed to be restricted to the lowest electron-and hole- superlattice subbands. The dashed lines are linear fits to $\rho(T)$. A quantitative analysis of the fitted slope versus the density is shown in Fig. 4c. Furthermore, an insulating behaviour near $\pm n_s$ is observed in electrical transport as T is increased above ≈ 90 K. To study the thermally activated transport behaviour of insulating states, we plot the temperature dependence of resistance at $\nu = \pm 4$ in Fig. S10(a),(b). An Arrhenius-like behaviour is clearly evident in this temperature range. From the slope in the Arrhenius plot, we estimate the activation gaps to be ~ 160 and ~ 240 K for the electron-side and hole-side respectively.

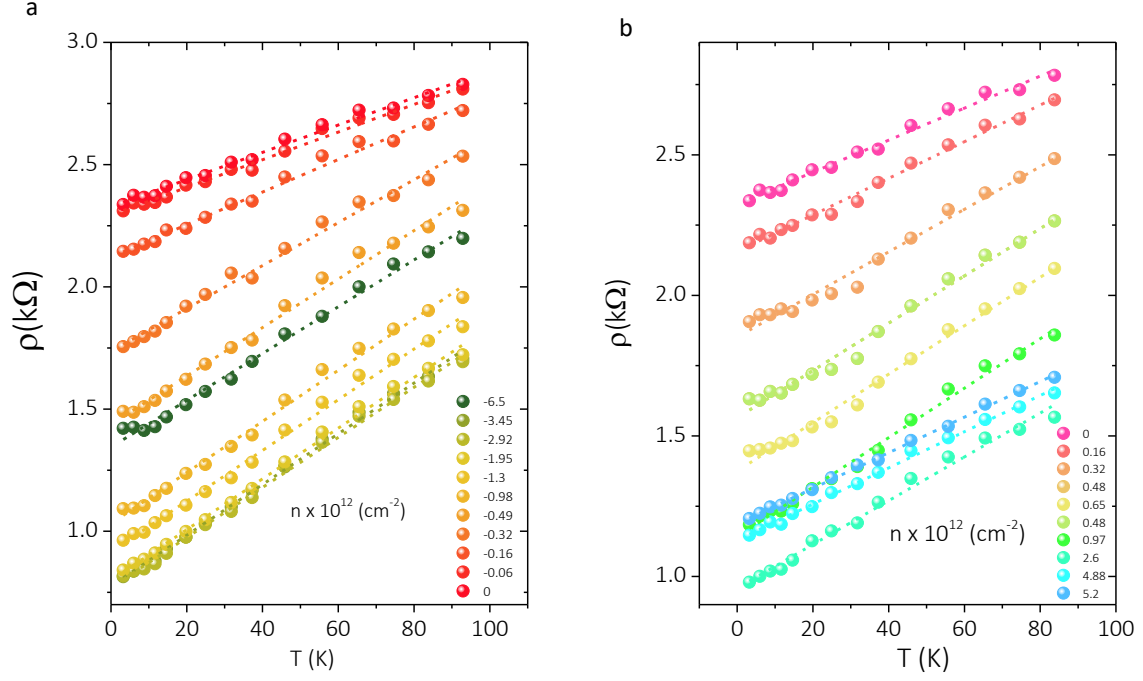


FIG. S9: (a),(b) ρ as a function of T for selected values of n in the T-linear regime. Dashed lines show linear fits to the data.

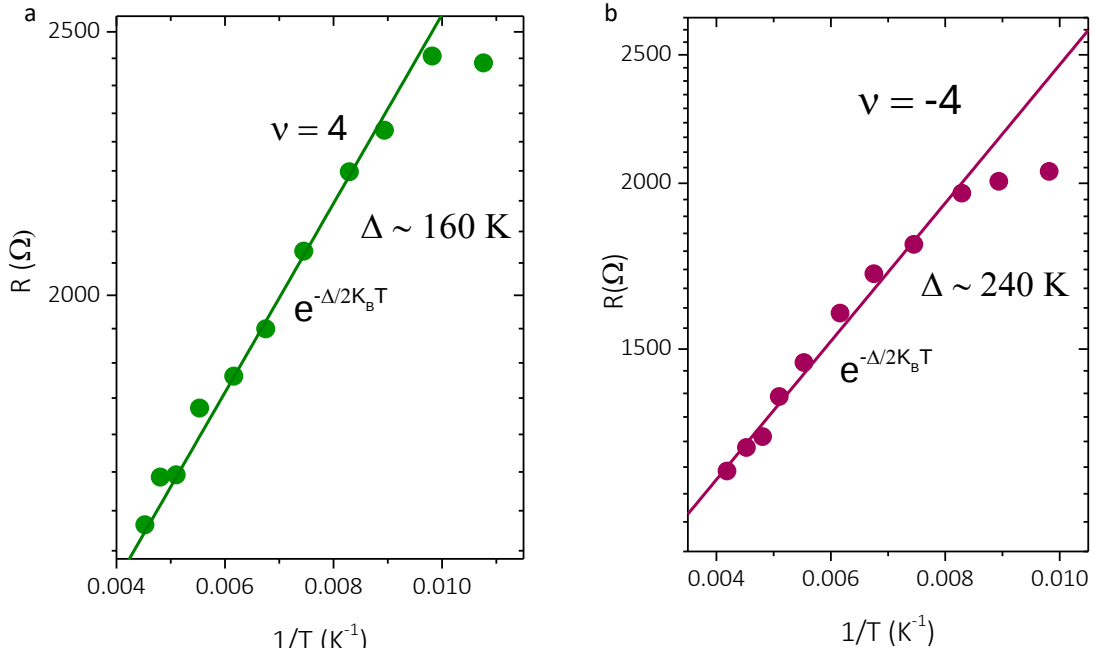


FIG. S10: (a),(b) R as a function of $1/T$ plotted for (a) $\nu = 4$, (b) $\nu = -4$. Solid lines show the Arrhenius-like fits to the data.

F. T-linear resistivity in D1 and D2 devices

Fig. S11(a), (b) show the linear slope of resistivity versus density for 1.24° and 1.01° respectively. Notably, we have restricted our analysis of $d\rho/dT$ to $\nu = \pm 3$, since at higher densities the insulating states at $\nu = \pm 4$ start influencing $\rho(T)$. Additionally, shaded regions in the two graphs correspond to regions which show insulating behaviour at low T . We find that the D5 device exhibits higher values of $d\rho/dT$ as compared to D4, which is consistent with previous studies, where $d\rho/dT$ reaches a maximum value closer to magic angle [11, 12]. Notably, in both the devices $d\rho/dT$ attains a maximum value near $\nu = -3$ and shows a clear e-h asymmetry.

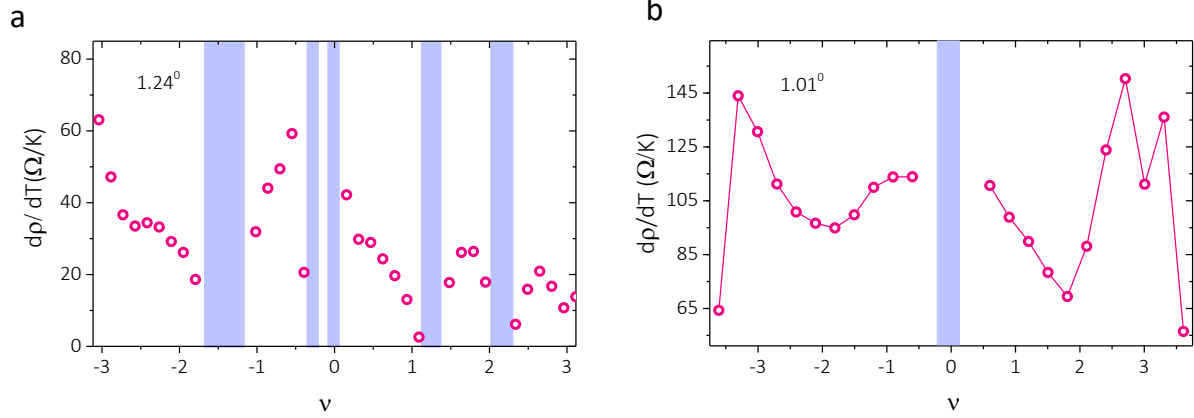


FIG. S11: $d\rho/dT$ extracted in the T-linear regime for (a) 1.24° and (b) 1.01° devices. The shaded regions represent the insulating regions observed in the two devices.

IV. Thermoelectric (TE) measurements in two different device geometries

We have carried out thermopower measurements in two different configurations depending on the device geometry, although the basic idea remains the same. For geometry C1, the measurements are schematically explained in Fig. S12(a). Briefly, a sinusoidal current (I_ω) is allowed to flow between two contacts (e.g. 1 and 2) of the monolayer branch outside the top gated region, setting up a temperature gradient (ΔT) across the tBLG region. The resulting second-harmonic thermo-voltage ($V_{2\omega}$) is recorded between leads 3 and 4 as a function of doping and heating current.[4, 13]. The thermopower results presented in main text for device D3 are obtained in this configuration, additionally this geometry allows us to measure thermopower in different configurations which are shown in section V. Similarly for H1 geometry, SLG region is used to create a (ΔT) across the tBLG channel, however in this case the voltage probes directly contact the tBLG channel as shown in Fig. S12(b). We find that our thermopower results are consistent in two geometries and hence are intrinsic to tBLG.

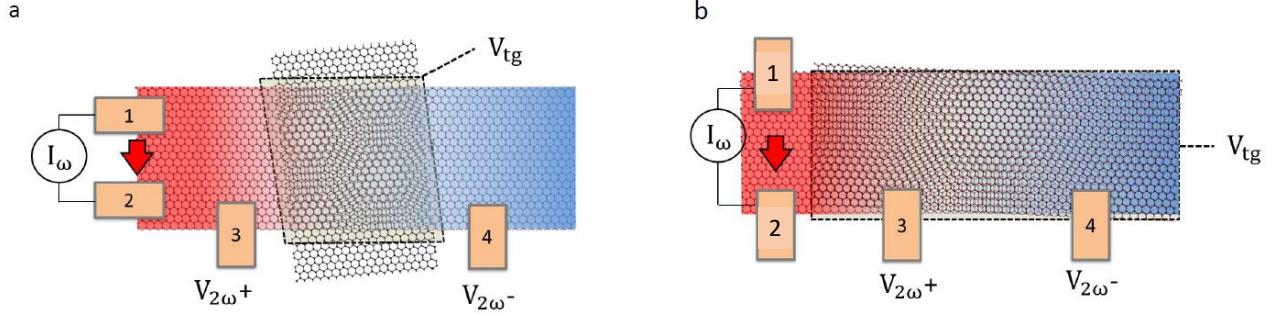


FIG. S12: Measurement schematic for thermo-voltage $V_{2\omega}$ in (a) C1 and (b) H1 device geometries.

V. Additional data on thermoelectric transport in different devices

A. Thermoelectric transport in device D2

Fig. S13(a) shows the density dependence of $V_{2\omega}$ measured for different heating currents. The linear response was ensured from $V_{2\omega} \propto I_\omega^2$ for the range of heating current used (Fig. S13(b)). The evolution of $V_{2\omega}$ with n is explained in Fig. 2 (main text). In brief, the system shows a sequence of CI (near $\nu = 1$ and 2) and CS (near $\nu = 3$) states as the Fermi energy approaches integer fillings, and correspondingly $V_{2\omega}$ exhibits a cascade of sign reversals or thermovoltage oscillations. (Shaded light blue regions near $\nu = 1, 2$ and 3). We note that, the h-side does not show a clear sequence of thermopower oscillations at integer fillings, which further highlights the e-h asymmetry present in the system. In order to connect the resistive features and thermopower, we compare the scaled $V_{2\omega}$ with $\alpha = (1/R)dR/dn$ in Fig. S13(b). Notably, while α qualitatively captures the sign reversals near integer fillings on the e-side, it fails to predict the excess thermopower near $\nu = 0.5$ (light pink shaded region), where $\alpha \approx 0$, which indicates a breakdown of the Mott formula as it is proportional to α . In addition, we observe a similar peak near $\nu = -1$ as well, which is also not captured by α , however the magnitude of excess thermopower is very less compared to the e-side.

In Fig. S13(c) we make a qualitative comparison between the measured thermopower and the calculated values from the Mott formula (S_{Mott}), where we have used the calculated DOS for 1.24° to compute S_{Mott} . We find that S_{Mott} predicts much higher values of thermopower for $1 < |\nu| < 2$ as compared to the measured $V_{2\omega}$. The observed behaviour is likely to originate from the fact that the DOS used is calculated from single-particle tight-binding approach, where e-e correlations are not included and hence does not represent the actual DOS in the device. Furthermore, to see the effect of temperature on thermopower, we show the normalized $V_{2\omega}$ and the corresponding α for three different temperatures in Fig. S13(d),(e) and (f). We find that the excess thermopower near $\nu = 0.5$ persists down to 280 mK and the qualitative behaviour of thermopower does not change as we go down in T .

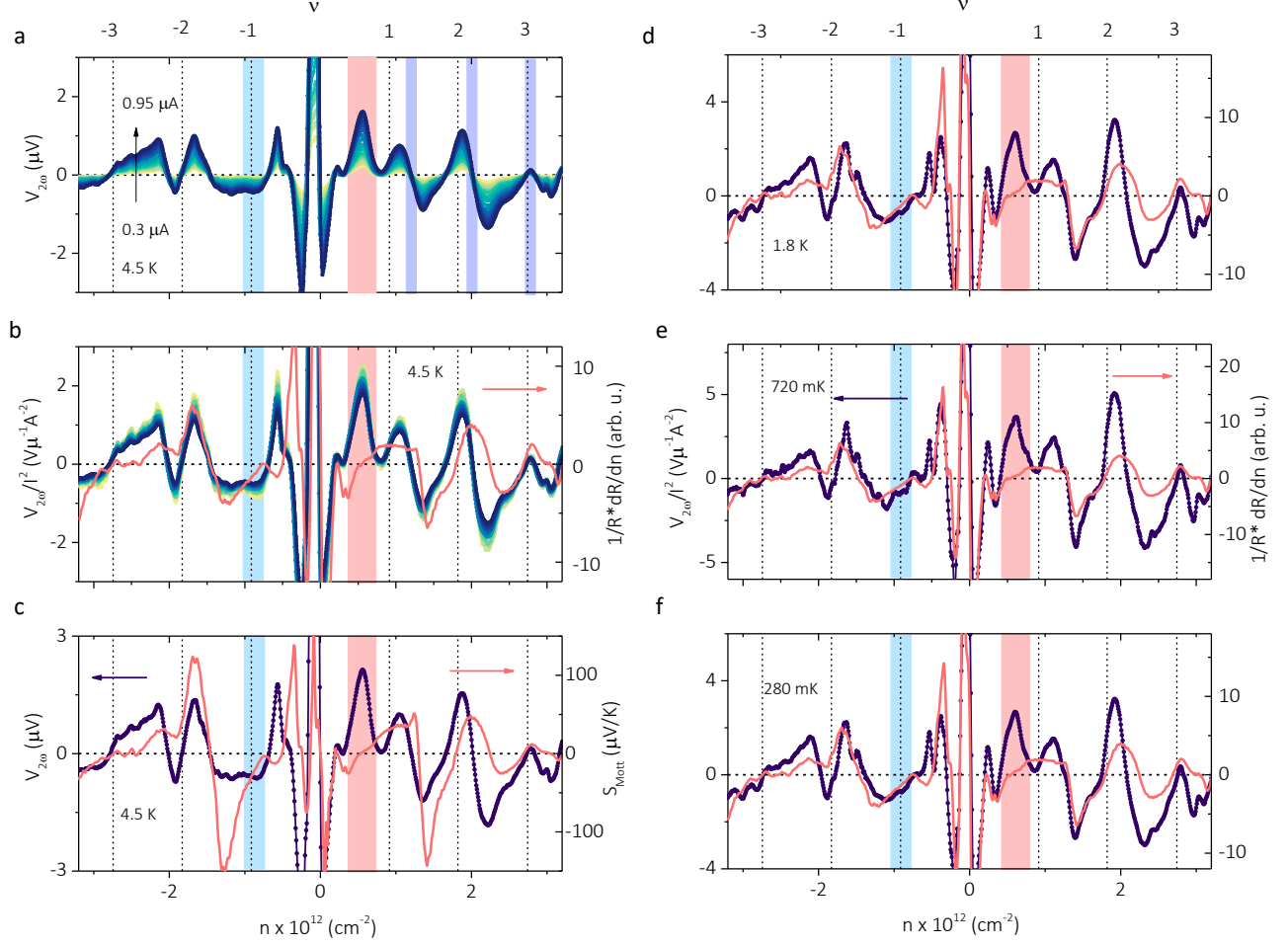


FIG. S13: (a) $V_{2\omega}$ as a function of n for different heating currents ($0.4 - 0.9 \mu\text{A}$) at 4.5 K . (b) $V_{2\omega}$ normalized with I_{ω}^2 at 4.5 K . The right axis in (b) shows the numerically calculated $(1/R)dR/dn$. (c) Comparison between the measured $V_{2\omega}$ (Left axis) and that calculated (right axis) from the semiclassical Mott relation (Eq. 2 in main text). (d) (e) (f): Comparison of $V_{2\omega}$ (left axis) with corresponding $(1/R)dR/dn$ (right axis) at 1.8 K , 720 mK and 280 mK respectively. Dashed lines in all graphs show the integer fillings. Light orange (cyan) shaded region represents excess thermopower on e(h) side, whereas light blue regions in (a) represent the gapped/semimetallic phases.

Thermoelectric transport in device D2 at higher temperature

The thermoelectric measurements shown in Fig. S13 were performed in a wet dilution refrigerator, where the maximum accessible T was 4.5 K . In order to probe the behaviour of excess thermopower and CI/CS states at higher T , the device was measured in a different cryostat in another thermal cycle. We note that the device characteristics degraded in this cycle; however, most of the features were reproduced qualitatively. Fig. S14 shows the thermopower data measured at five different temperatures, where $V_{2\omega}$ is normalized with respect to the CNP in order to focus on the excess thermopower and the correlated CI/CS phases. We note that the excess thermopower near $\nu = 0.5$ (albeit very weak compared to the previous thermal cycle) starts vanishing around $10 - 12 \text{ K}$. On the contrary, we find that the CI/CS phases persist upto $20 - 25 \text{ K}$, pointing to a different physical mechanism responsible for the two features even though both originate from e-e interactions in the system. However, further understanding requires theoretical considerations and more experiments.

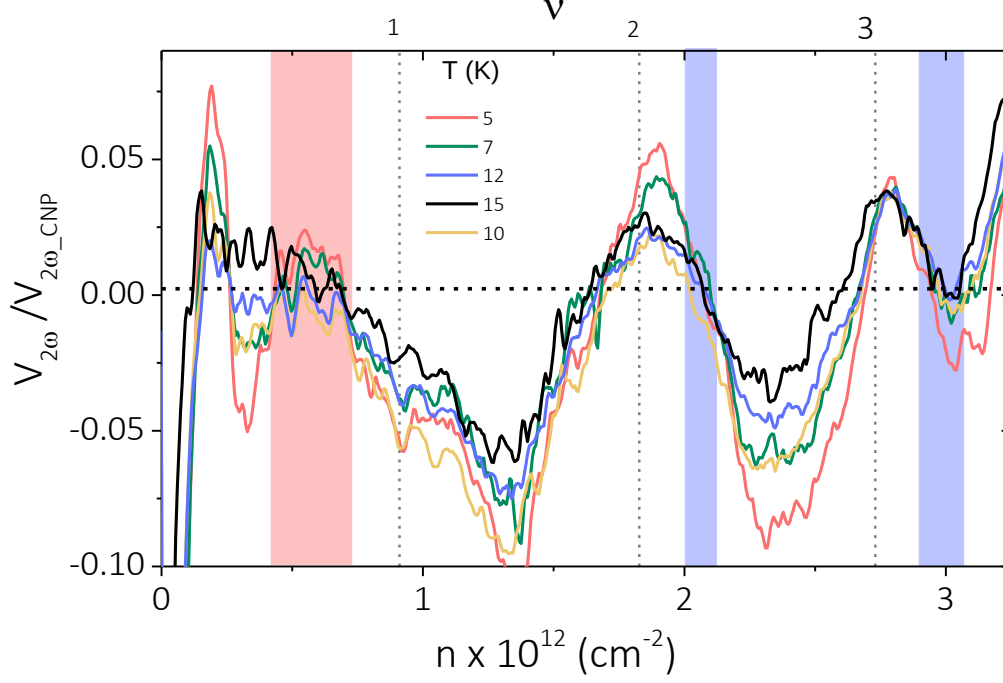


FIG. S14: $V_{2\omega}$ as a function of n for five different temperatures. The CI/CS states are marked by light blue shaded regions, whereas the light pink region highlights the excess theropower peak.

B. Seebeck coefficient fitted with Mott formula in device D3

The fitting of the Mott formula (S_{Mott}) obtained using the calculated density of states (DOS) for three different twist angles with the measured thermopower for the 1.6° device is depicted in Fig. S15. We observe that S_{Mott} from the DOS for 1.61° matches well with measured $V_{2\omega}$ both at the CNP and $\nu = \pm 4$ simultaneously, whereas if the twist angle is tuned away from 1.6° , we find a deviation in the Mott fit at $\nu = \pm 4$ as clearly shown for 1.69° and 1.41° . This further emphasises that the thermoelectric transport is extremely sensitive to the band structure and in turn any correlation effects, which can not be probed with conductance measurements alone.

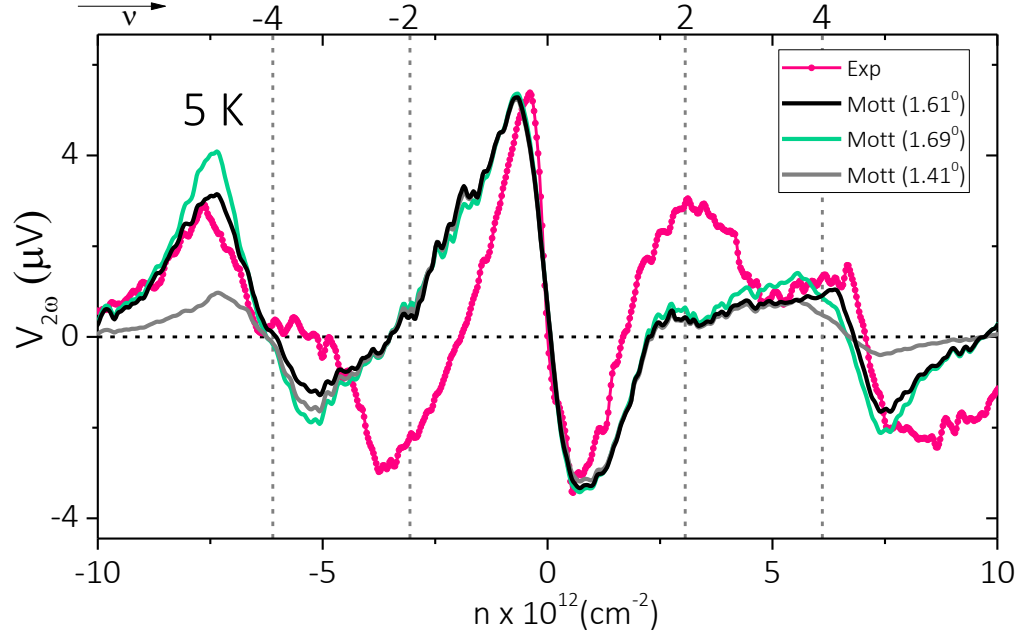


FIG. S15: Doping dependence of the measured $V_{2\omega}$ compared with S_{Mott} calculated using DOS for three different twist angles 1.61° , 1.69° and 1.41° .

Seebeck coefficient fitted with Mott formula at various temperatures in device D3

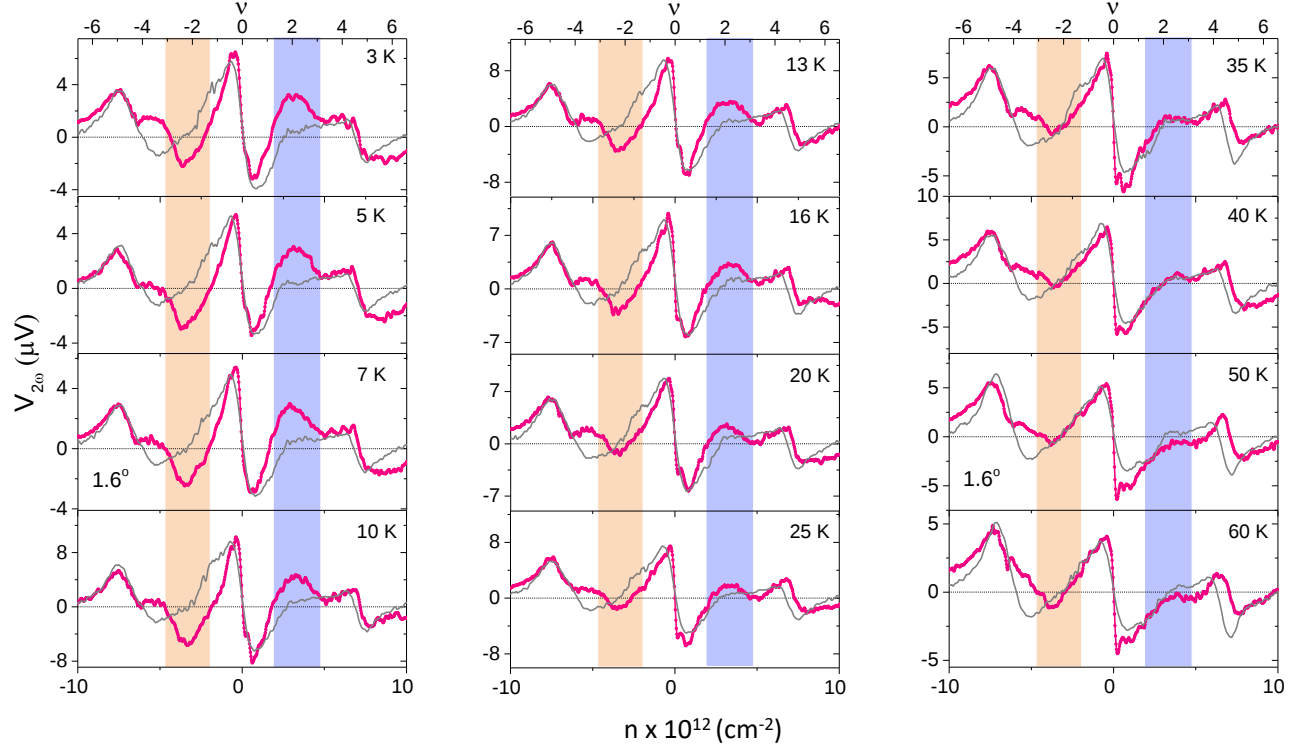


FIG. S16: Comparison between the measured $V_{2\omega}$ (pink lines) and that calculated (Grey line) from the semiclassical Mott relation at different temperatures.

C. ΔT calibration for device D3

The thermoelectric power (TEP) or Seebeck coefficient S is obtained from the ratio of thermoelectric voltage and the temperature gradient ($V_{2\omega}/\Delta T$) across the tBLG. In our earlier work on tBLG [1, 4], we have used graphene resistance thermometry for obtaining ΔT , which is not particularly effective at our device geometry since the 2-probe resistance of the tBLG regions cannot be accurately calibrated to the temperature of the tBLG channel. Hence we employ Mott formula to estimate ΔT . This method of obtaining ΔT relies on the assumption that the system follows the Mott formalism, which is indeed true for low angle tBLG [1]. Furthermore, the Mott formula exhibits an excellent match with the experimental data both at the CNP and $\nu = \pm 4$ simultaneously (Fig. 3b), which in turn justifies our method. We have fit the Mott formula to $V_{2\omega}$ at different values of T and obtained ΔT as shown in Fig. S17(a). Additionally, as evident from the Fig. S17(b), (c) and (d) the obtained Seebeck coefficient shows T-linear dependence in the dispersive band as expected from the T-linear dependence from Mott relation, further validating the employed method.

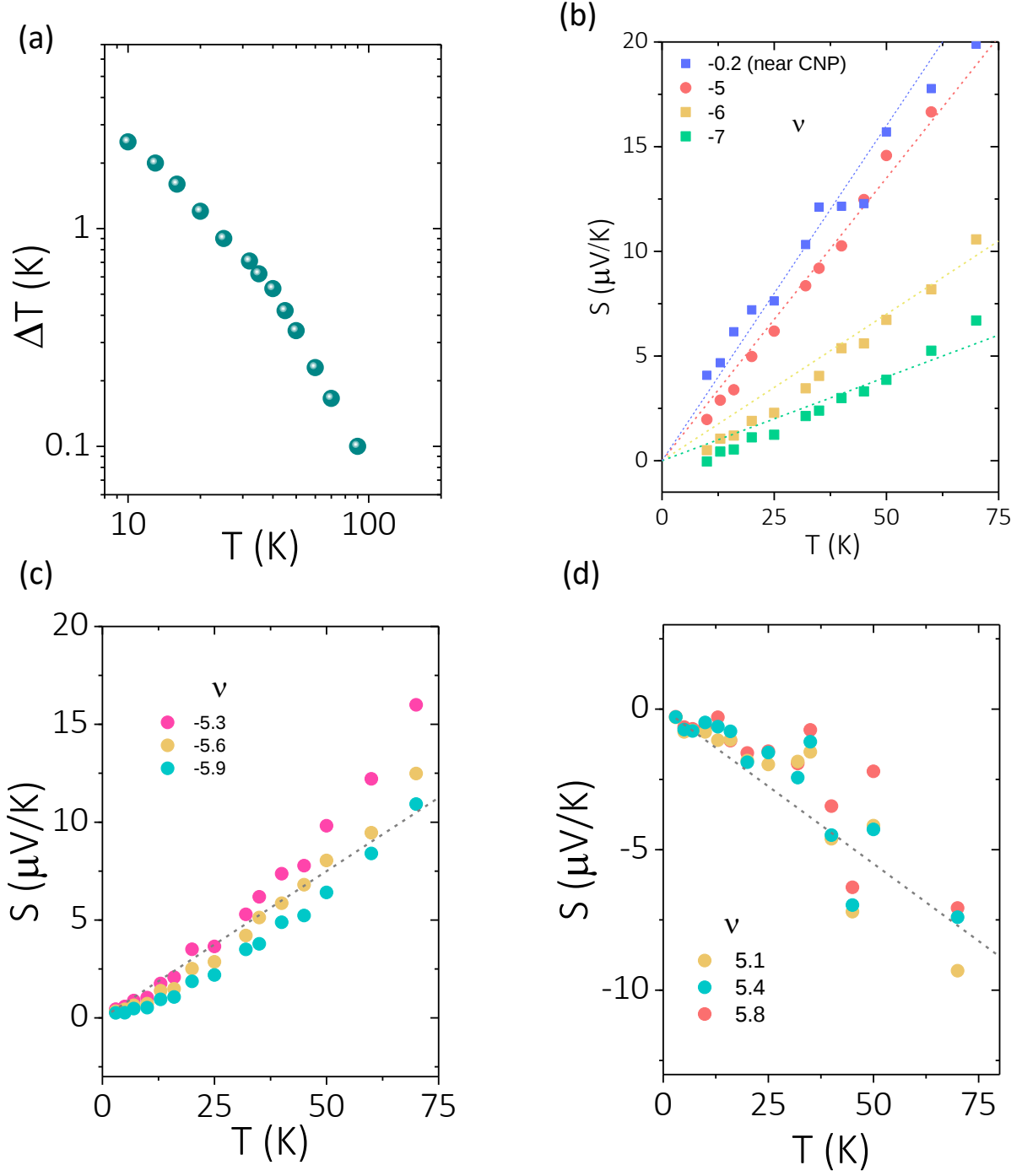


FIG. S17: (a) The extracted values of ΔT obtained from Mott fit at various T . (b),(c) and (d) S as a function of T at CNP and at different fillings in higher dispersive bands. Dashed lines show the T-linear behaviour as a guide for eye.

D. Estimation of T_F for device D3 using tight-binding model

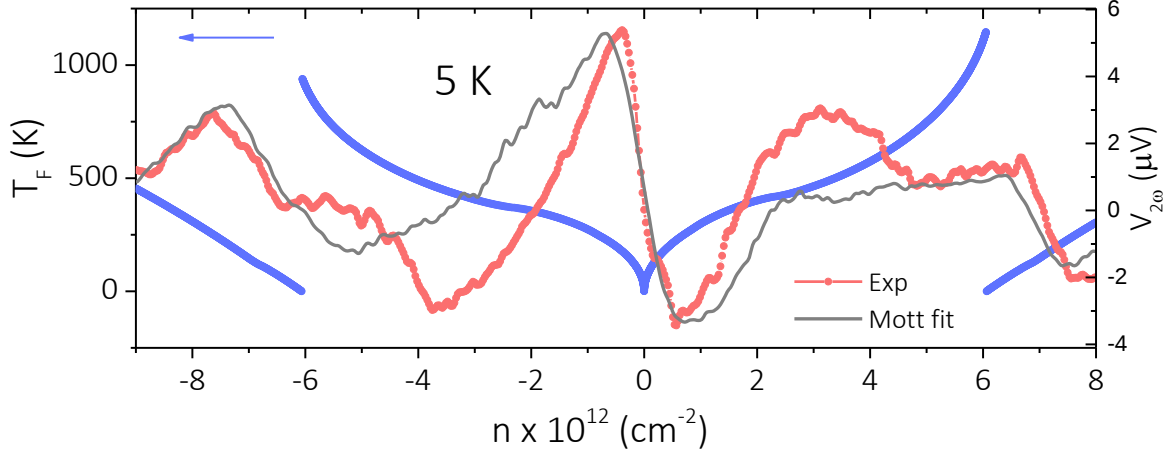


FIG. S18: Doping dependence of T_F (left axis) calculated using tight-binding model plotted with the measured $V_{2\omega}$ and calculated thermopower using Mott formula (right axis). The Fermi temperature (T_F) is obtained from the band structure calculation by converting the energy scale to temperature. However, we have explicitly assumed that the effective number density of the conducting carriers is re-normalized to zero at $\nu = \pm 4$ due to the band gap. Consequently, T_F goes to zero at $\nu = \pm 4$ since the effective number density is zero. While fitting at the local maxima/minima of $V_{2\omega}$ near CNP ($n \approx 0.6 \times 10^{12} \text{ cm}^{-2}$), the value of $T_F \approx 240 \text{ K}$. Similarly, near the local maxima close to $|\nu| = 4$ ($n \approx 7.6 \times 10^{12} \text{ cm}^{-2}$), the value of $T_F \approx 250 \text{ K}$ and satisfies the degenerate limit $T \ll T_F$ for fitting the Mott formula.

E. Doping dependence of $S - S_{\text{Mott}}$ for device D3

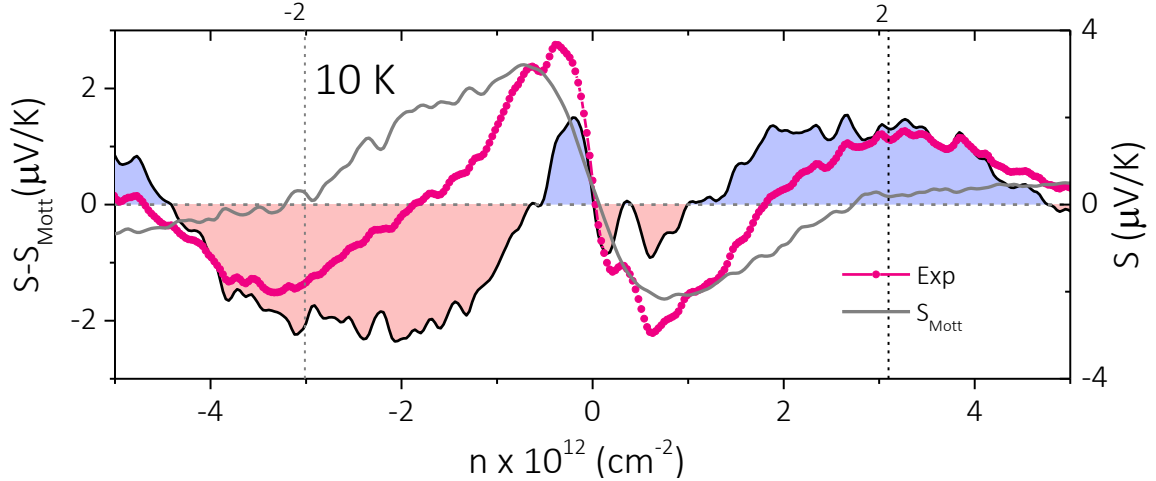


FIG. S19: Doping dependence of measured $S - S_{\text{Mott}}$ at 10 K, which attains maximum value near $\nu = 2$ on the e-side and between $\nu = -1$ and -2 on the h-side, reflecting the e-h asymmetry present in the system.

F. Thermoelectric measurements in parallel magnetic field for device D3

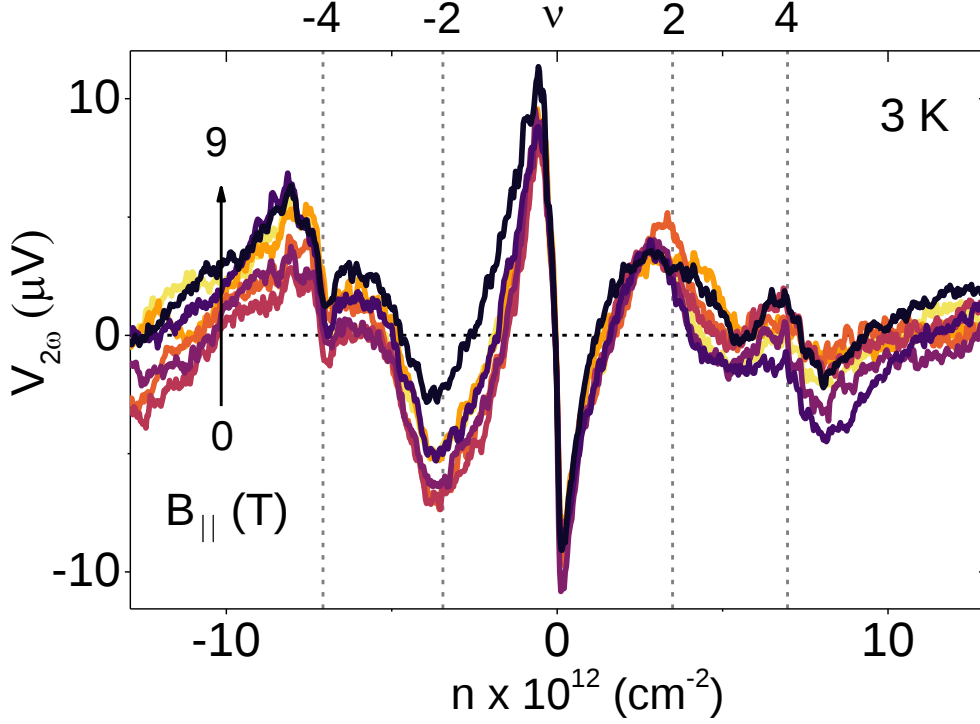


FIG. S20: Thermopower $V_{2\omega}$ as a function of n for different magnetic fields applied parallel to plane of the tBLG measured at 3 K. No appreciable change in $V_{2\omega}$ is observed for magnetic fields upto 9 T.

G. Thermoelectric measurements in different heating configurations in device D3

We have performed thermoelectric measurements in different heating configurations to verify the results described in Fig. 3. The optical micrograph of the device along with the contact configuration is shown in Fig. S21(a). Briefly, a sinusoidal current is passed between two contacts to create a ΔT in the tBLG region and the resulting second harmonic voltage $V_{2\omega}$ is measured. Fig. S21(b) shows the TE measured in three different configurations at 5 K. We note that apart from the sign reversal at the CNP and the vHS, two new extrema, consisting of a maximum close to $\nu = +2$ and minimum near $\nu = -2$ develop, which is very similar to the results described in main text (Fig. 3b). However, a slight asymmetry in different configurations is observed, which can arise from local inhomogeneity in the twist angle. Furthermore, we have performed temperature dependent TE measurements in the configuration given by, $I : 1 - 9, V : 3 - 4$. Fig. S22 shows $V_{2\omega}$ for six different temperatures. We note that as T is increased above 35 – 40 K, the excess TE near $\nu = \pm 2$ vanishes, indicating the T scale upto which correlation effects are present. Reproducibility of our TE results in different heating configurations ascertains the distinct nature of the ground state.

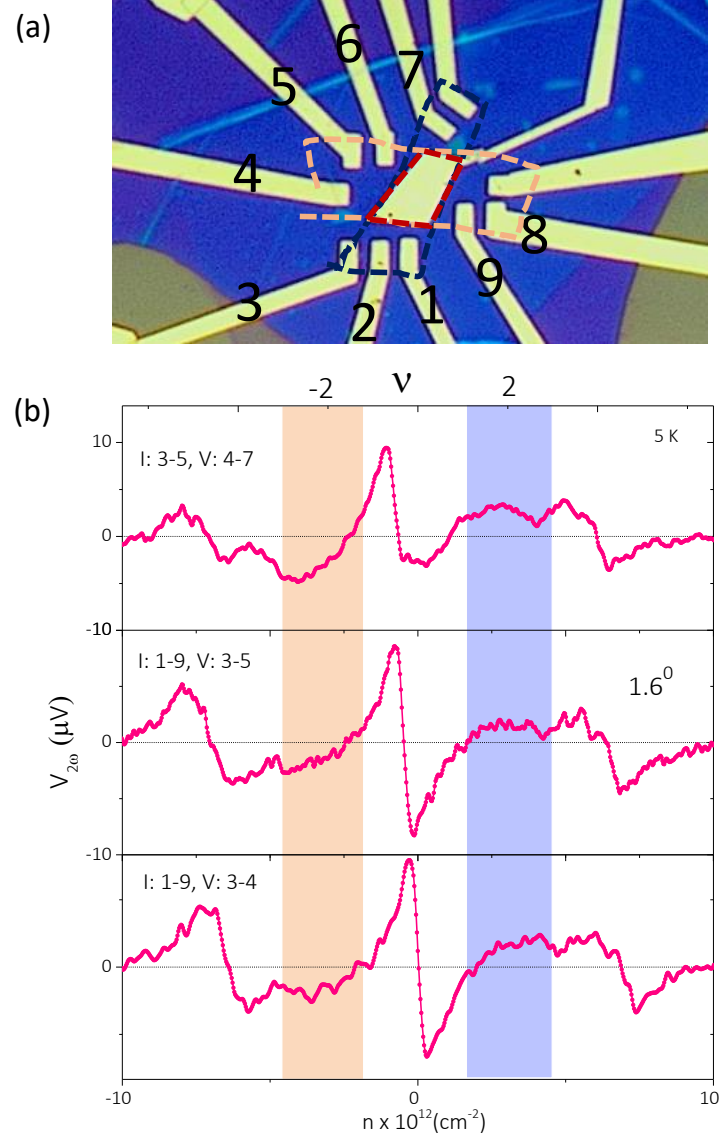


FIG. S21: (a) Optical micrograph of the device with the contact configuration. (b) Measured $V_{2\omega}$ for three different heating configurations.

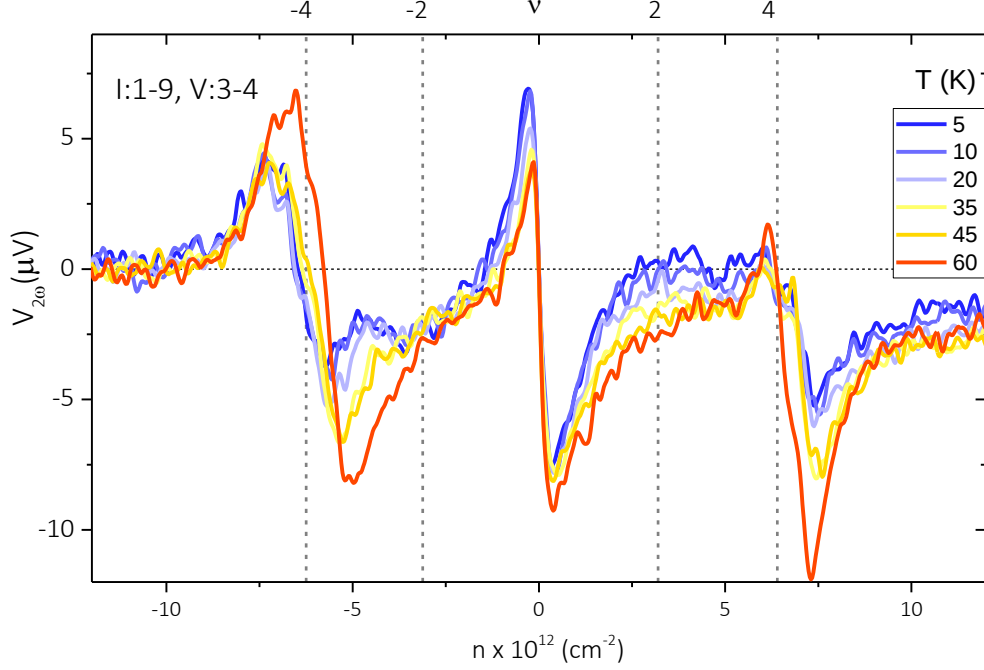


FIG. S22: Measured $V_{2\omega}$ for six representative T in the configuration $I : 1 - 9, V : 3 - 4$. Dashed lines show filling factors.

VI. Planckian dissipation

In non-Fermi Liquid state (NFL), the incoherent scattering is often found to be bound by the Planckian dissipation scale $\tau^{-1} = Ck_B T/\hbar$, where the dimensionless coefficient C is order of unity.

$$C = \frac{\hbar}{k_B} \frac{e^2 n_c}{m^*(0)} A \quad (S1)$$

where n_c is the gate-induced carrier density and $m^*(0)$ is cyclotron mass at $T \rightarrow 0$ which is proportional to the density of states per Fermi pocket at the Fermi energy for two dimensional materials. We calculate the effective mass (m^*) as a function of n in the devices with twist angles 1.6° , 1.7° and 4° using the expression, $m^* = (\hbar^2/2\pi)D(E)/N$, where N is the degeneracy. We have used $N=8$ (as evident from the Landau fan) and the density of states as calculated by tight binding method for 1.6° , 1.7° and the SLG DOS for 4° . Fig. S23(a),(b) show the calculated m^* for the three devices. We note that our calculated values of m^* for 1.6° matches well with previously measured value for a device with similar twist angle [11]. The above expression allows us to calculate C and hence the scattering rate τ^{-1} over a range of number density as shown in Fig. 4d. We find that $C \sim O(1)$ for $\theta \approx 1.6^\circ$ and 1.7° near $\nu = \pm 2$, where the deviation from Mott relation is maximum. In comparison, the value of C is two orders of magnitude smaller for $\theta \sim 4^\circ$.

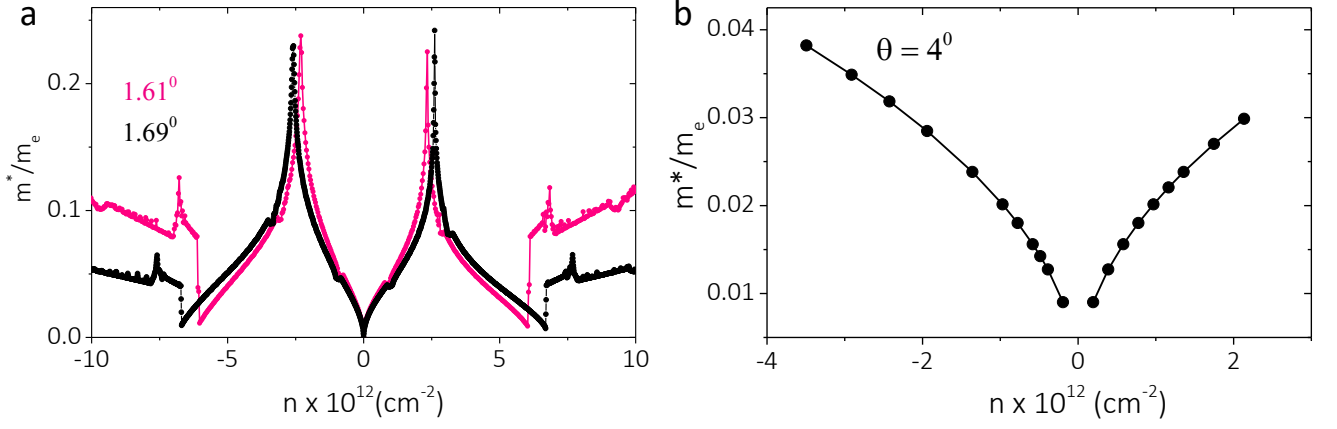


FIG. S23: Numerically calculated cyclotron mass for (a) $\theta = 1.61^\circ, 1.69^\circ$ and (b) $\theta = 4^\circ$

VII. Theoretical calculations

A. Computational Details for the tight binding formalism

The electronic hamiltonian of the system is written as

$$\hat{\mathbf{H}} = - \sum_{i,j} t(\mathbf{R}_i - \mathbf{R}_j) c_i^\dagger c_j + \text{h.c.} = - \sum_{i,j} t_{ij} c_i^\dagger c_j + \text{h.c.} \quad (\text{S2})$$

where $\mathbf{R}_i$ denotes the real space position of the i^{th} atom, and $c_i^\dagger$ and c_i are the creation and annihilation operators at $\mathbf{R}_i$. We approximate the transfer integrals t_{ij} using the Slater Koster formalism [14] assuming that the overlap of the p_z orbitals can be approximated as the linear combination of the $\sigma\sigma$ and the $\pi\pi$ overlaps. Taking the local curvature of the sheets into account the transfer integral can be written as [15]:

$$t_{ij} = t_{\pi\pi} [\hat{\mathbf{n}}_i - (\hat{\mathbf{n}}_i \cdot \hat{\mathbf{R}}_{ij}) \hat{\mathbf{R}}_{ij}] \cdot [\hat{\mathbf{n}}_j - (\hat{\mathbf{n}}_j \cdot \hat{\mathbf{R}}_{ij}) \hat{\mathbf{R}}_{ij}] + t_{\sigma\sigma} [\hat{\mathbf{n}}_i \cdot \hat{\mathbf{R}}_{ij}] [\hat{\mathbf{n}}_j \cdot \hat{\mathbf{R}}_{ij}] \quad (\text{S3})$$

where $\hat{\mathbf{n}}_i$ is the unit normal at the i^{th} site, and $\hat{\mathbf{R}}_{ij}$ is the unit vector joining the sites.

We can see that from fig.(S24) that if there is no local curvature, and:

- i. if i and j are on the same layer then the only non-zero contribution is from the $\pi\pi$ term
- ii. if i and j are on different layers then $t_{ij} = t_{\sigma\sigma} \cos^2(\theta) + t_{\pi\pi} \sin^2(\theta)$ where θ is the angle that $\hat{\mathbf{n}}_i$ makes with $\hat{\mathbf{R}}_{ij}$

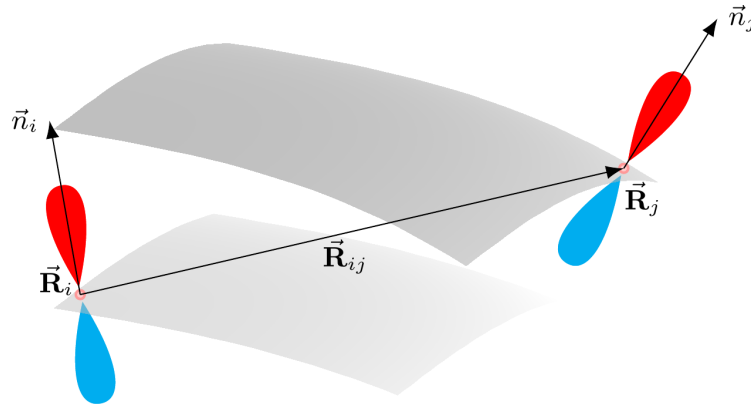


FIG. S24: Local normals at $\mathbf{R}_i$ and $\mathbf{R}_j$. Note that, $\hat{\mathbf{n}}_{i/j} = \frac{\vec{n}_{i/j}}{|\vec{n}_{i/j}|}$ and $\hat{\mathbf{R}}_{ij} = \frac{\vec{R}_{ij}}{|\vec{R}_{ij}|}$

The terms $t_{\pi\pi}$ and $t_{\sigma\sigma}$ are taken as follows:

$$t_{\pi\pi} = t_{\pi}^0 \exp\left(-\frac{|\vec{\mathbf{R}}_{ij}| - a_0}{\delta}\right); \quad t_{\sigma\sigma} = t_{\sigma}^0 \exp\left(-\frac{|\vec{\mathbf{R}}_{ij}| - d_0}{\delta}\right) \quad (\text{S4})$$

On setting the parameter $a_0 = 2.45 \text{ \AA}$, the nearest neighbour distance between two Carbon atoms, it is reasonable to take $t_{\pi}^0 \approx -2.7 \text{ eV}$, the nearest neighbour transfer energy in monolayer graphene. Similarly by taking $d_0 = 3.35 \text{ \AA}$, the interlayer distance in AA stacked bilayer, we fix $t_{\sigma}^0 \approx 0.48 \text{ eV}$. The attenuating factor, $\delta = 0.184a_0$ is chosen such that the strength of the next nearest neighbour is 0.1 times the nearest neighbour interaction in monolayer graphene.[16]. To compute the density of states we use an analog of the linear tetrahedron method for 2D systems, the linear triangulation method. A uniform (110×110) grid is taken in the Brillouin Zone (BZ) and a delaunay triangulation is performed on those points. The density of states and the number density are then estimated by integrating within each of the triangles.

B. Lattice model for the low-energy bands

To set up the dynamical mean field theory (DMFT) calculations involving the four low-energy bands near the CNP we assume an effective hexagonal lattice model for the moire lattice with two electronic orbitals and two spins ($\sigma = \pm 1/2$) indexed by $\alpha = 1, \dots, \mathcal{M}$ with $\mathcal{M} = 4$, such that there are four bands that can hold a maximum of eight electrons per triangular unit cell, i.e.

$$\mathcal{H} = - \sum_{ij,\alpha} t_{ij} c_{i\alpha}^{\dagger} c_{j\alpha} + \mathcal{H}_{int} \quad (\text{S5})$$

Here $c_{i\alpha}$ is the electron operator for i -th hexagonal lattice site. The hopping integrals are in general complex and can be chosen to fit [17–19] the energy dispersion from band-structure calculation, e.g. that shown in Fig.1d of the main text. However, the two-orbital model is known [18, 20] to be insufficient to reproduce the topology of the Bloch wave functions while keeping all the low-energy symmetries of the continuum model [21]. Within the simplified DMFT approximation discussed below only the DOS of the low-energy bands enter and we take the DOS directly from our full tight-binding bandstructure calculation discussed in the main text. Hence the tight-binding parameterization for the low-energy effective lattice model does not explicitly appear in our DMFT calculations. We also neglect the asymmetry between the orbitals induced by the band structure. The interaction term $\mathcal{H}_{int} = U \sum_{\square} (\sum_{i \in \square, \alpha} n_{i\alpha}/3 - 4)^2$ is the cluster Hubbard term [17, 18] where the sum runs over all the hexagons in the moire lattice and $n_{i\alpha}$ is the electron number operator for α orbital/spin on the i -th site. The cluster term arises since the physical charges are concentrated around the AA regions of the moire lattice. When expanded the cluster Hubbard term leads to on-site as well as longer-range interactions with strength comparable to the on-site one [17]. The longer-range terms could be important for various possible symmetry-breaking orders [17, 18, 22, 23] at low temperature near magic angles. Finding the phase diagram of the cluster Hubbard model for arbitrary filling is an interesting, but challenging theoretical problem [22, 23].

However, for the metallic state considered here within DMFT, the extended terms are mainly expected to lead to screening and renormalization of the on-site Hubbard U [24]. To this end, we take

$$\mathcal{H}_{int} = U \sum_{i, \alpha < \gamma} n_{i\alpha} n_{i\gamma}, \quad (\text{S6})$$

i.e. an $SU(4)$ -symmetric on-site Hubbard interaction. The minimal model already captures the crucial effects of interaction near the low-energy VHS, as discussed below and in the main text. The strength of the on-site U could be estimated as $U = 1.857(e^2/4\pi\epsilon\epsilon_0 L_M)$ [17, 25], where the moire lattice constant $L_M = a_0/(2\sin(\theta/2)) = 8.89 \text{ nm}$ for graphene lattice spacing $a_0 = 0.246 \text{ nm}$ and twist angle $\theta = 1.6^\circ$. Hence $U \simeq 15 - 75 \text{ meV}$ depending on the dielectric constant $\epsilon \simeq 20 - 4$ or from screening due to the gates. Based on our estimated total band width of $W = 180 \text{ meV}$ for the four low-energy bands near CNP, we get a moderate interaction strength $U/W \simeq 0.1 - 0.4$. For our DMFT calculations, we take $U/W \simeq 0.2$. We find that the effect of this moderate interaction gets enhanced near the low-energy van Hove singularities (VHS).

C. Dynamical mean field theory

In the DMFT approximation [26], and assuming a homogeneous state with orbital and spin ($SU(4)$) symmetry, we reduce the model of Eq.(S5) to an effective single-site impurity problem with the imaginary-time action

$$S_{imp} = - \int_0^\beta d\tau d\tau' \sum_\alpha \mathcal{G}^{-1}(\tau - \tau') \bar{c}_\alpha(\tau) c_\alpha(\tau') + U \sum_{\alpha < \gamma} \int_0^\beta d\tau n_\alpha(\tau) n_\gamma(\tau) \quad (S7)$$

Here $\beta = 1/k_B T$ and $(\bar{c}_\alpha, c_\alpha)$ are fermionic Grassmann variables with $n_\alpha = \bar{c}_\alpha c_\alpha$. The dynamical mean field $\mathcal{G}^{-1}(i\omega_n) = i\omega_n + \mu - \Delta(i\omega_n)$ for the Matsubara frequency $\omega_n = (2n + 1)k_B T$, with n being an integer, is determined by the hybridization function $\Delta(i\omega_n)$ which is self-consistently determined using the non-interacting lattice DOS as we discuss below. The chemical potential is fixed by the filling.

Since we need the real-frequency electronic Green's function $G(\omega) = G(i\omega_n \rightarrow \omega + i0^+)$ to compute dc transport coefficients, e.g. the Seebeck coefficient, we use an approximate impurity solver, namely modified iterative perturbation theory (IPT) [26, 27] and its generalization for the multi-orbital case [28]. The latter has been benchmarked with numerically exact techniques for solving the single-impurity problem [28], and is expected to work quite well for moderate interaction strengths, like in our case. Within the IPT, we obtain the impurity self-energy as

$$\Sigma(\omega) = (\mathcal{M} - 1)U\langle n \rangle + \frac{A(\mathcal{M} - 1)\Sigma^{(2)}(\omega)}{1 - B(\mathcal{M} - 1)\Sigma^{(2)}(\omega)}, \quad (S8)$$

and the impurity Green's function is obtained from the Dyson equation $G^{-1}(\omega) = \mathcal{G}^{-1}(\omega) - \Sigma(\omega)$. The first term in Eq.(S8) is the Hartree self energy and $\Sigma^{(2)}(\tau) = U^2 \tilde{\mathcal{G}}^3(\tau)$ is the second-order self-energy obtained using the Hartree-corrected impurity Green's function $\tilde{\mathcal{G}}^{-1}(\omega) = \mathcal{G}^{-1}(\omega) - U\langle n \rangle$. The coefficients A and B are chosen to satisfy certain sum rules and the known high-frequency behaviour of the impurity Green's function [28] and are given by

$$A = \frac{\langle n \rangle (1 - \langle n \rangle)}{\langle n_0 \rangle (1 - \langle n_0 \rangle)} + \frac{(\mathcal{M} - 2)(\langle nn \rangle - \langle n \rangle^2)}{\langle n_0 \rangle (1 - \langle n_0 \rangle)} \quad (S9)$$

$$B = \frac{(1 - (\mathcal{M} - 1)U\langle n \rangle) + \mu_0 - \mu}{(\mathcal{M} - 1)U^2 \langle n_0 \rangle (1 - \langle n_0 \rangle)} \quad (S10)$$

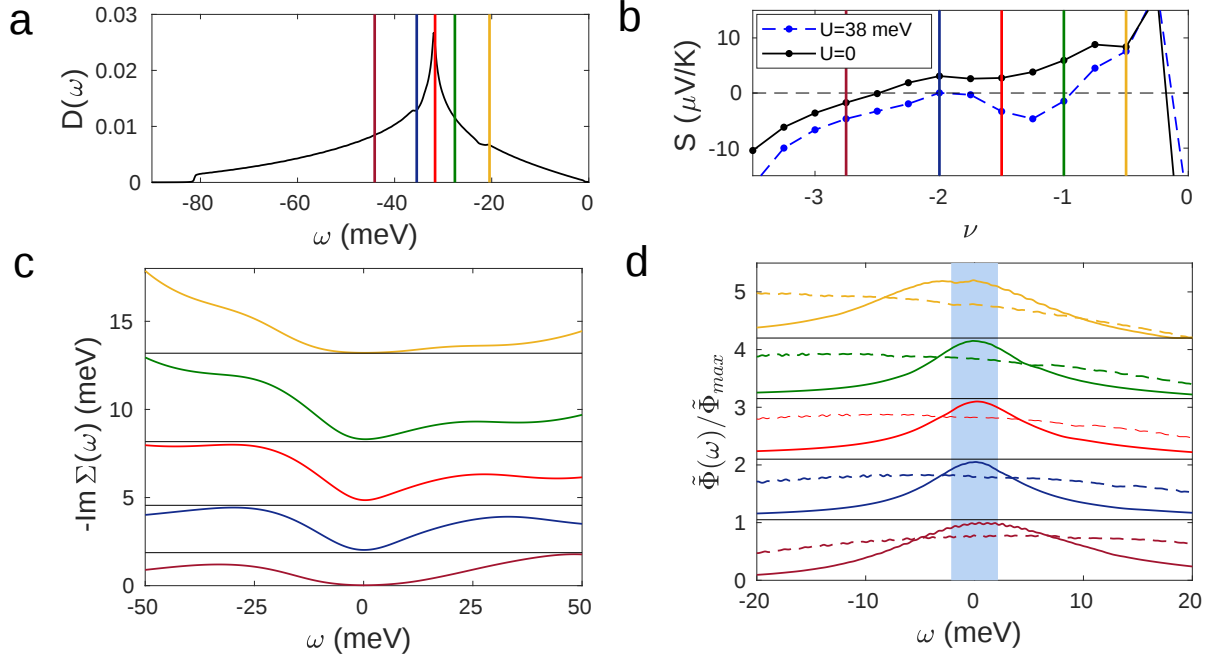


FIG. S25: **Van Hove singularity, self-energy and the sign of thermopower** : (a) Non-interacting DOS from the tight binding bandstructure calculation for densities (or chemical potentials) below the CNP for $T = 14$ K. The vertical lines indicate five densities across the low-energy van Hove singularity (VHS). (b) The same densities or fillings (ν) are indicated by the vertical lines for the Seebeck coefficient (S) vs. ν plot at $T = 14$ K for the both non-interacting ($U = 0$, black line with filled circles) and the interacting ($U = 38$ meV, blue lines with filled circles) cases. The sign changes for the interacting and non-interacting cases happen at different fillings. (c) Imaginary part of electronic self energy at $T = 14$ K for the five fillings shown in (a) and (b) as indicated by the colors. For clarity, the self-energies at the subsequent fillings are vertically shifted by the baselines indicated by the solid horizontal lines. The electronic self-energy is enhanced near the VHS with marked low-energy particle-hole asymmetry. The Fermi liquid-like behavior, $-\text{Im}\Sigma(\omega) \sim \omega^2$, at low energies persist over a narrower energy window near the VHS indicating a more correlated metallic state with a lower coherence temperature scale. (d) Effective transport DOS $\tilde{\Phi}(\omega)$ (solid lines) for the same five fillings as in (a), (b) and (c), indicated by the same colors. Again, $\tilde{\Phi}(\omega)$ for the five subsequent fillings are vertically shifted for clarity. The large self-energy effects near the VHS strongly modify the effective transport DOS for the interacting case compared to the non-interacting $\tilde{\Phi}(\omega)$ (dashed lines). The shaded region has a width $\sim k_B T$ determined by $-T(\partial n_F(\omega)/\partial \omega)$ that controls the integral [Eq.(S13)] appearing in Eq.(S12) for the Seebeck coefficient. The interacting $\tilde{\Phi}(\omega)$ becomes sharply peaked near the VHS. The sign of S depends on whether $\tilde{\Phi}(\omega)$ increases or decreases with ω near $\omega = 0$ (see text for a discussion).

Here $\langle n \rangle = -(1/\pi) \int_{-\infty}^{\infty} d\omega n_F(\omega) \text{Im}G(\omega) = (\nu + 4)/8$ is the occupancy of a single orbital. The density-density correlation function $\langle nn \rangle \approx -\int_{-\infty}^{\infty} d\omega n_F(\omega) \text{Im}(\Sigma G)/(\pi U(\mathcal{M} - 1))$ with $n_F(\omega) = 1/(e^{\beta\omega} + 1)$, the Fermi function. Following ref.29, we fix the ‘pseudo’ chemical potential μ_0 from $\langle n_0 \rangle = -(1/\pi) \int_{-\infty}^{\infty} d\omega n_F(\omega) \text{Im}\tilde{G}(\omega) = \langle n \rangle$ using the Hartree corrected Green function $\tilde{G}^{-1}(\omega) = \omega + \mu_0 - \Delta(\omega) - U\langle n \rangle$.

Once the self-energy [Eq.(S8)] is obtained, the DMFT self-consistency condition is used to compute the local lattice Green’s function

$$G(\omega) = \frac{1}{2\mathcal{M}} \int_{-\infty}^{\infty} d\epsilon \frac{D(\epsilon)}{\omega + \mu - \epsilon - \Sigma(\omega)}, \quad (\text{S11})$$

assumed to be the same as the impurity Green’s function for a single orbital and spin species. Here $D(\epsilon)$ is the total

tight-binding DOS per triangular moire unit cell for the four lowest bands. The self-consistency loop is closed by obtaining the new hybridization function $\Delta(\omega) = \omega + \mu - \epsilon - \Sigma(\omega) - G^{-1}(\omega)$. The loop is iterated till we reach convergence.

We fix the interaction strength $U = 38$ meV and perform the calculations to obtain electronic Green's function and self-energy for fillings $-3 < \nu < 3$ and temperatures $5\text{K} \lesssim T \lesssim 100$ K. The Seebeck coefficient [30] S is given by,

$$S = -\frac{k_B}{e} \frac{A_1}{A_0} \quad (\text{S12})$$

where

$$A_m = \int_{-\infty}^{\infty} d\omega d\epsilon \rho^2(\epsilon, \omega) \Phi(\epsilon) \left(-T \frac{\partial n_F(\omega)}{\partial \omega} \right) (\beta\omega)^m \quad (\text{S13})$$

with $m = 1, 2$. $\rho(\epsilon, \omega) = -(1/\pi) \text{Im}(1/(\omega + \mu - \epsilon - \Sigma(\omega)))$ is obtained from DMFT self-energy and $\Phi(\epsilon) = (1/\mathcal{A}) \sum_{n=1, \mathbf{k}}^4 (\partial \epsilon_{n\mathbf{k}} / \partial k_x)^2 \delta(\epsilon - \epsilon_{n\mathbf{k}})$ is the transport DOS that is obtained from tight-binding energy dispersion $\epsilon_{n\mathbf{k}}$ for the four bands near CNP. Here $\mathcal{A}$ is the area of the sample. To analyze the sign of the Seebeck coefficient, we define an effective transport DOS, $\tilde{\Phi}(\omega) = \int_{-\infty}^{\infty} d\epsilon \rho^2(\epsilon, \omega) \Phi(\epsilon)$. In the non-interacting case ($U = 0$), we take $\rho(\epsilon, \omega) \sim \eta / ((\omega + \mu - \epsilon)^2 + \eta^2)$ with a small phenomenological broadening $\eta \simeq 0.001W$ mimicking the effect of impurity scattering. In this case, $\rho^2(\epsilon, \omega)$ is sharply peaked around $\omega = \epsilon - \mu$ and $\tilde{\Phi}(\epsilon, \omega)$ is effectively determined by the tight-binding transport DOS $\Phi(\epsilon)$. As a result, at low temperatures A_1, A_0 and hence S is determined by the behavior of $\Phi(\omega + \mu)$ over an energy window $\sim k_B T$ around the chemical potential ($\omega = 0$). Hence the sign of A_1 or the Seebeck coefficient is negative (positive) depending on whether $\Phi(\omega + \mu)$ increases (decreases) with ω , and $S \approx 0$ when $\omega = 0$ is at the peak of $\Phi(\omega + \mu)$. As we show in Figs.S25, for the interacting case treated within DMFT, due to the large and strongly temperature-dependent self-energy effects near the VHSs, $\tilde{\Phi}(\omega)$, unlike $\Phi(\omega + \mu)$, becomes very sharply peaked around the chemical potential and varies strongly over energy window $\sim k_B T$. These lead to the non-standard sign and the violation of SMR over a range $-2 \lesssim \nu \lesssim -1$ as shown in Fig. 4(e) and Fig. S26(a).

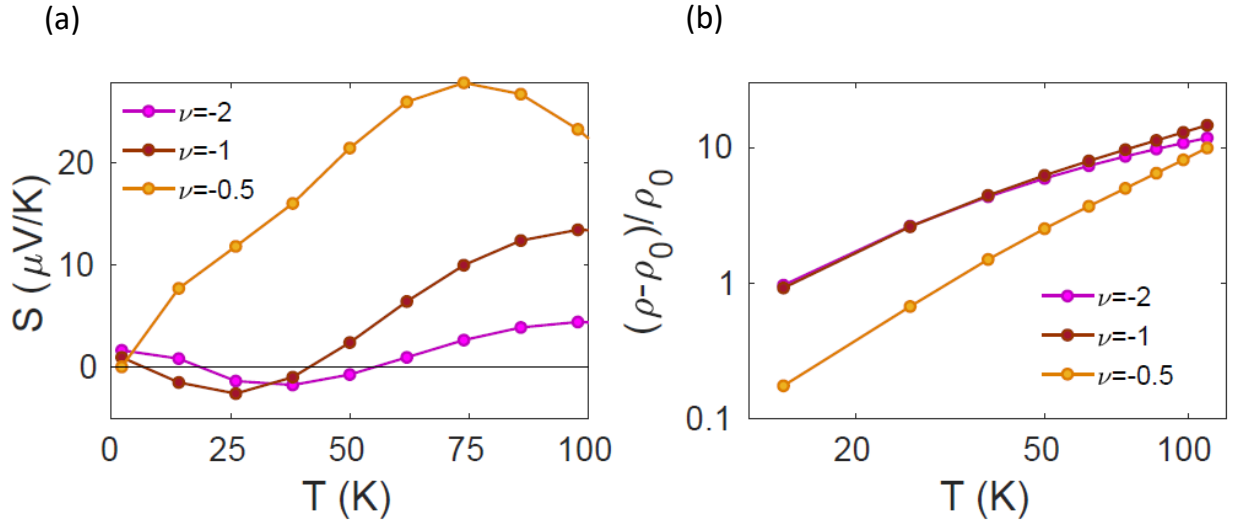


FIG. S26: (a) Computed S from DMFT as a function of temperature for fillings $\nu = -2, -1$ and -0.5 . The Seebeck coefficient changes from positive to negative sign over an intermediate temperature range. (b) Normalized resistivity ρ computed from DMFT as a function of T for fillings $\nu = -2, -1$ and -0.5 , where ρ_0 is the residual resistivity. The dashed line shows T -linear dependence.

As shown in Fig. S25, we also find strong particle-hole asymmetry in the self-energy and DOS even at low-energies near $\omega = 0$ (chemical potential). The particle-hole asymmetry results from the particle-hole asymmetry of the non-interacting DOS at intermediate energies away from the VHSs, as evident in Fig.3a. The asymmetry is further

enhanced by the interaction. This asymmetry also influences the sign of the thermopower near the VHSs at low and intermediate temperatures. We also obtain a linear- T resistivity (Fig. S26(b)) over an extended temperature range. This is, of course, a known outcome of DMFT for correlated systems [31, 32]. However, to obtain the detailed density dependence and the magnitude of resistivity one needs to consider the electron-phonon scattering and the effects of long-range impurities relevant at such low-densities near CNP [33, 34]. The thermopower and the violation of SMR are not expected to be influenced significantly by electron-phonon and impurity scattering [35, 36].

D. Violation of Mott relation in DMFT

In Fig.S27(d), we show the deviation $\Delta S = S - S_{\text{Mott}}$ from the Mott value S_{Mott} (Fig.S27(b)) obtained from resistivity ρ (Fig.S27(a)) computed in DMFT using the Mott formula of Eq.1 of the main text. The computed ΔS (Fig. 1d) is large in the intermediate temperatures ($< U/k_B$), indicating strong violation of Mott law in DMFT, in contrast to that at low temperature. However, a comparison of this violation of the Mott law in DMFT to the experimentally observed violation of SMR is somewhat misleading, since the former mainly arises from the strong filling dependence of the DMFT resistivity, which exhibits additional broad peaks near $|\nu| = 2$, due to large electron-electron (el-el) scattering near the van Hove singularity (vHS). Such additional peaks are absent in the experimentally measured resistivity (Fig.1, main text), though seen during thermal cycles (Fig. S8). Due to these additional resistivity peaks, the computed S_{Mott} in DMFT is large at intermediate temperatures and has additional sign changes (Fig.S27(b)) compared to the actual thermopower S (Fig.S27(c)), leading to large ΔS i.e. strong violation of Mott law (Fig.S27(d)).

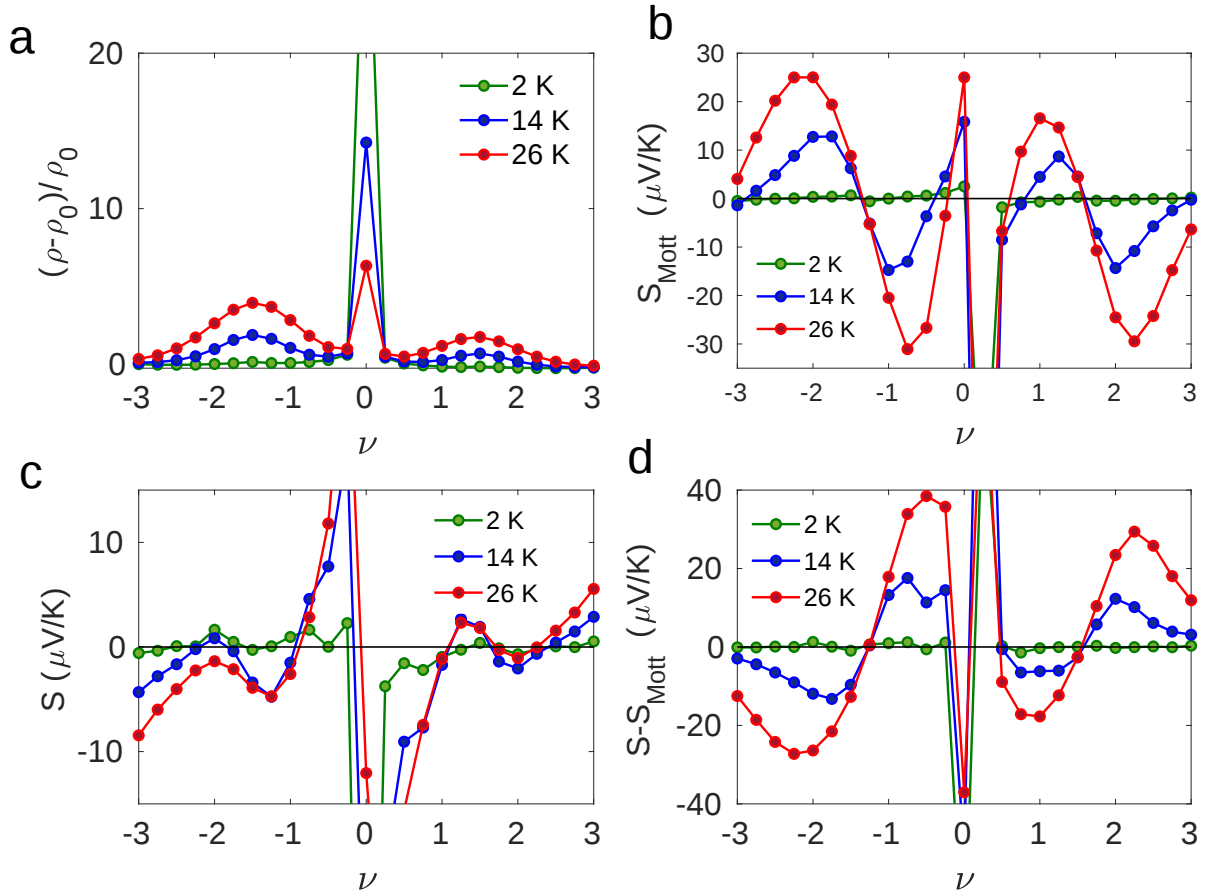


FIG. S27: (a) Normalized resistivity $(\rho - \rho_0)/\rho_0$ relative to the residual resistivity ρ_0 as a function of ν for $T = 2, 14, 26$ K. (b) $S_{\text{Mott}}(\nu)$ computed in DMFT using Mott law. (c) Thermopower S computed from DMFT. (d) The deviation $\Delta S = S - S_{\text{Mott}}$ from Mott value in DMFT.

We would also like to emphasize that our DMFT calculations only capture intrinsic el-el contributions to resistivity and thermopower, and impurity scatterings, which leads to the residual resistivity ρ_0 , is only treated in an approximate manner in terms of a filling independent impurity broadening in the DMFT Green's functions. Hence our calculations cannot capture the large residual resistance seen in experiment and its filling dependence, presumably arising from the details of the impurity scattering and its possible intricate interplay with el-el scattering at low-temperature. These effects are beyond the scope of our current theoretical calculations. However, as is well known [35], scatterings from static impurities alone, even if large, cannot give rise to violation of the Mott law. The excess thermopower in the DMFT calculations could be quantified better by comparing with the non-interacting results at $U = 0$ as shown in Figs.S28(a),(b). Here, we can clearly deduce the excess thermopower and its opposite sign appearing close to $|\nu| = 2$ due to the enhanced el-el interaction near the vHSs.

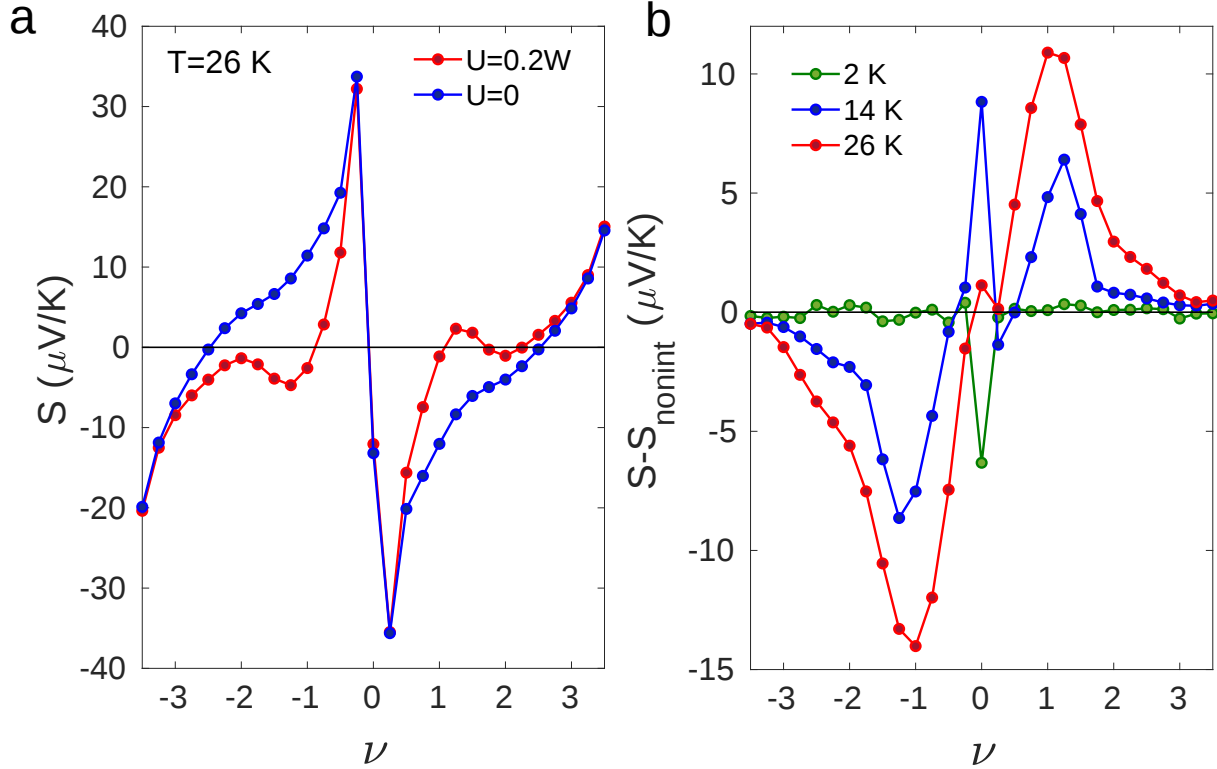


FIG. S28: (a) Comparison of thermopower for interacting ($U = 0.2W$) and non-interacting ($U = 0$) cases at $T = 26$ K. (b) The excess thermopower $S - S_{nonint}$ due to el-el interaction is large for $|\nu| \simeq 1 - 2$ near the vHSs.

E. Comparison between excess thermopower and entropy in DMFT

The connection between thermopower and thermodynamic entropy (S_{ent}) is typically understood in terms of so-called Kelvin formula [37, 38], $S = -(1/|e|)(\partial S_{ent}/\partial n)$, where n is the number density of the charge carrier. The Kelvin formula can be obtained for non-interacting electrons from the SMR (Eq.1, main text) by assuming that the only dependence of the resistance R on energy E is due to density of states, and the velocity and scattering time are energy independent. Hence the Kelvin formula is only an approximation. However, the formula works quite well for several strongly-correlated systems [37, 39], and the formula is found to be exact in certain models of non-Fermi liquid metals [38, 40] that are dual to charged black holes [41]. We do not find the Kelvin formula to describe our results for DMFT thermopower S well, presumably due to strong energy dependence of velocity due to the vHSs. However,

as shown in Fig.S29, we find the excess entropy due to el-el interactio, over the non-interacting ($U = 0$) entropy, to correlate well with the excess thermopower (Fig.S28) as a function of filling and temperature. We compute the entropy (per hexagonal moire lattice site, spin and orbital) in DMFT using the Maxwell relation $(\partial S_{ent}/\partial n)_T = -(\partial \mu/\partial T)_n$, where $\mu(T)$ is obtained in DMFT by fixing the filling. We obtain the $S_{ent}(\nu, T)$ by integrating over $(\partial S_{ent}/\partial n)_T$ from $\nu = -4(4)$ to $\nu < 0(> 0)$, assuming $S_{ent}(|\nu| = 4, T) = 0$ i.e. when the low-energy band is completely empty or full. This is justified in our theoretical model which does not consider the higher bands.

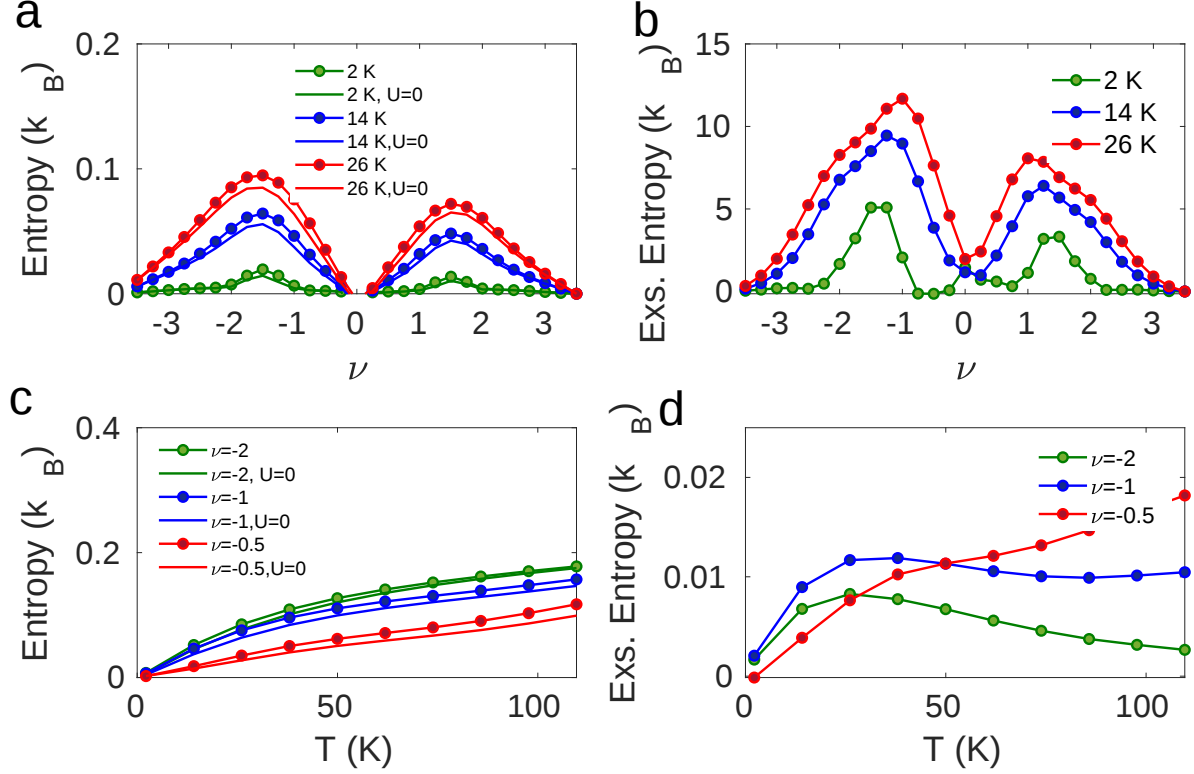


FIG. S29: (a) Comparison of entropy S_{ent} as a function of filling ν for $T = 2, 14$ and 26 K for interacting ($U = 0.2W$) and non-interacting ($U = 0$) cases. (b) The excess entropy $S_{ent}(U = 0.2W) - S_{ent}(U = 0)$ (in units of $10^{-3}k_B$) as a function of ν . (c) Comparison of S_{ent} for interacting and non-interacting cases as a functions of temperature for $\nu = -2, -1, -0.5$ and (d) corresponding excess entropies as function of temperature.

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