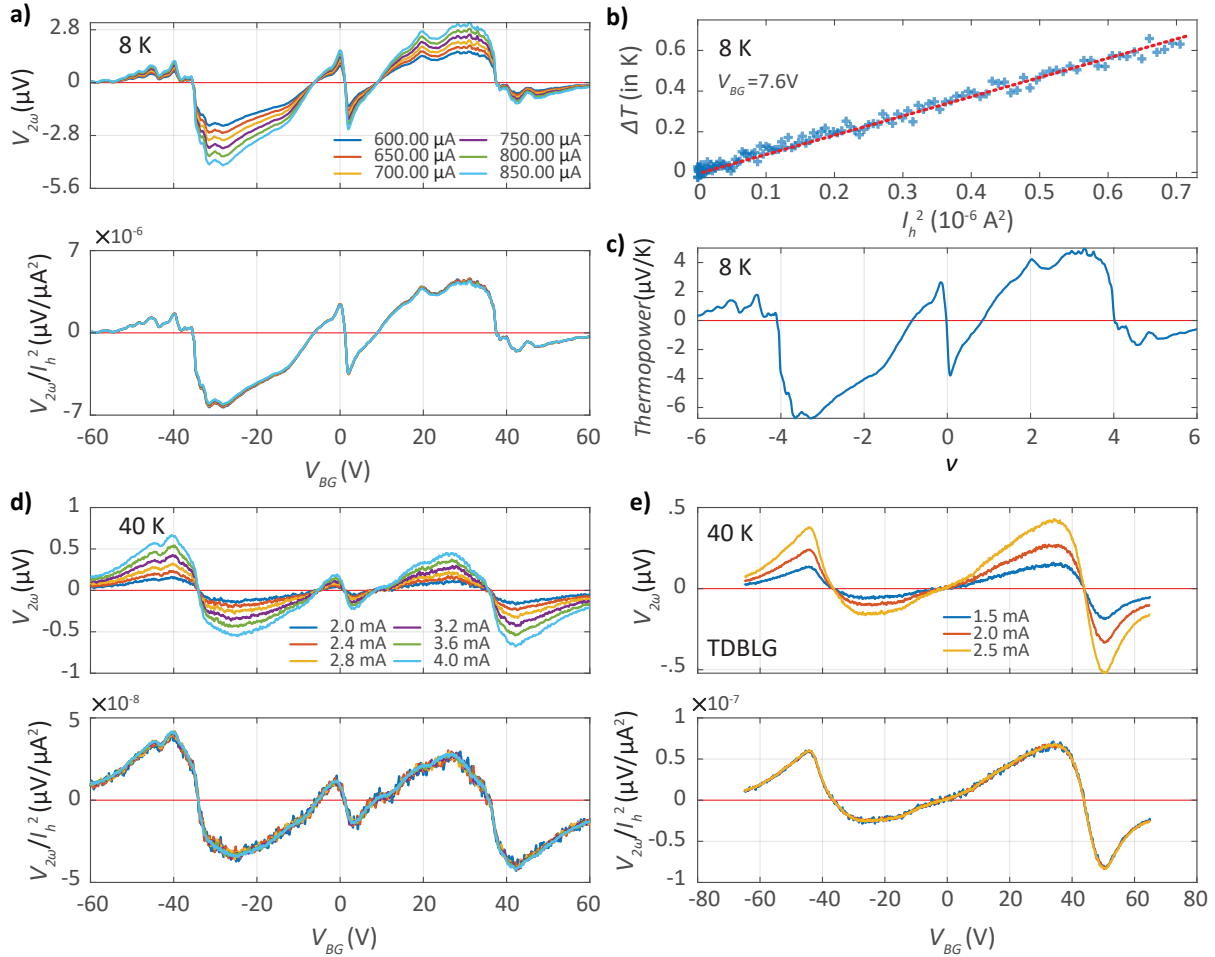
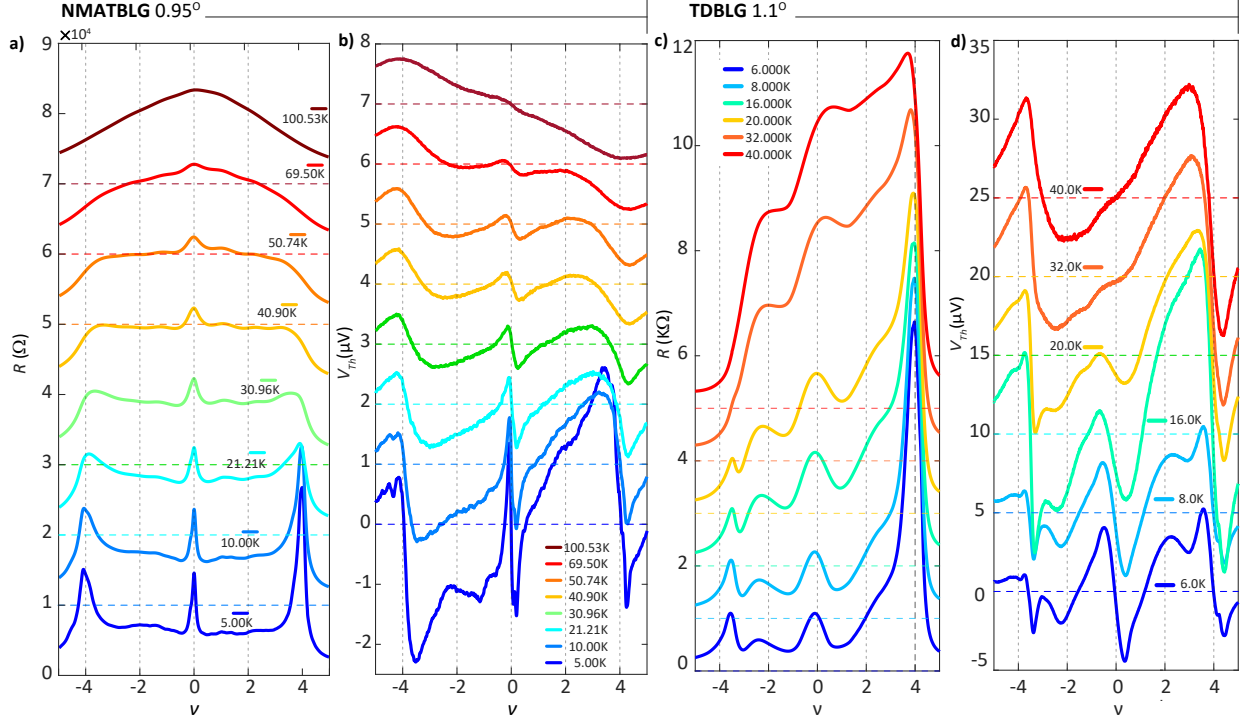


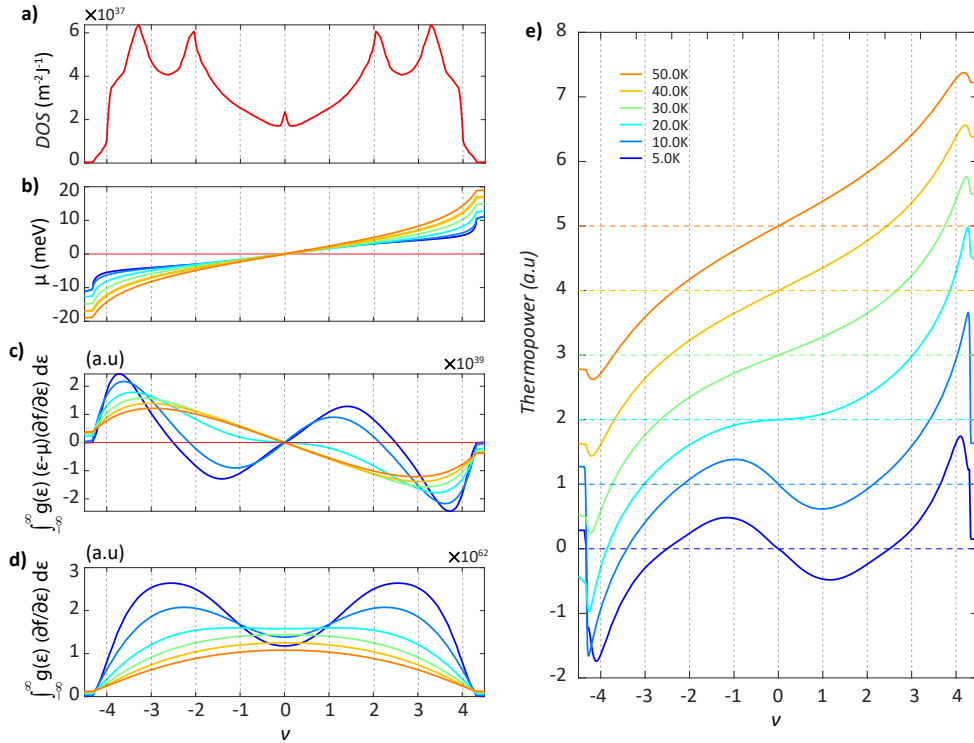
**Extended Data Fig. 1: Resistance response as a function of density ( $n$ ): (a), (b), and (c),** Resistance as a function of density for MATBLG (at 5K), near-MATBLG (at 5K) and TDBLG (at 4K), respectively. For all three devices, we have used the full-filling ( $\nu = \pm 4$ ) carrier density ( $n_s$ ) to measure the twist angle ( $\theta$ ) using  $n_s = 8\theta^2/\sqrt{3}a^2$ , where  $a = 0.246\text{nm}$  is the lattice constant of monolayer graphene. For MATBLG, near-MATBLG and TDBLG  $n_s \approx 2.58 \times 10^{12}$ ,  $2.15 \times 10^{12}$  and  $3.19 \times 10^{12}\text{cm}^{-2}$  respectively, which translates to  $\theta \sim 1.05^\circ$ ,  $0.95^\circ$  and  $1.1^\circ$  respectively. Inset shows the optical images of the corresponding devices. The scale bars are  $5\mu\text{m}$ .



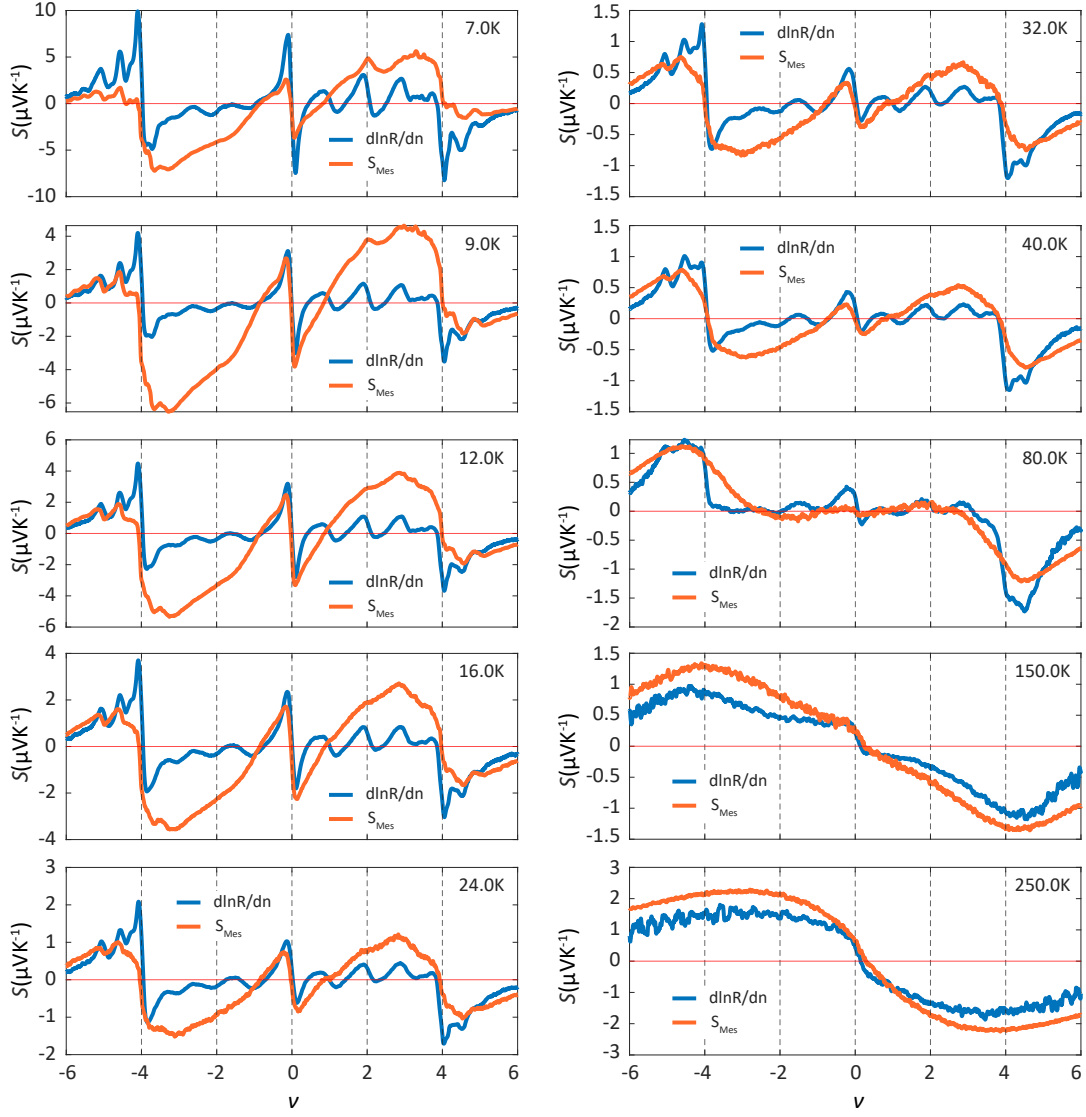
**Extended Data Fig. 2: The linear regime of thermopower measurement:** (a)  $V_{2\omega}$  signal versus  $V_{bg}$  for different applied heater currents ( $I_h$ ), respectively, at 8K for MATBLG device (top panel).  $V_{2\omega}$  signal normalized by the square of respective heater currents ( $I_h^2$ ) versus backgate voltage  $V_{bg}$  (bottom panel). All the normalized  $V_{2\omega}/I_h^2$  values overlap well showing the  $V_{2\omega}$  measurements have been performed in the linear response regime. (b) Measured temperature difference ( $\Delta T$ ) as a function of  $I_h^2$  at 8K at backgate voltage  $V_{bg} = 7.6V$ . Within the maximum heater current range, a clear linear trend (dashed red line) can be observed between  $\Delta T$  and  $I_h^2$ , and  $\Delta T \ll T$ . (c) Thermopower as a function of filling  $\nu$  derived from measured  $V_{2\omega}$  signal and  $\Delta T$ . (d), (e) (Top panels)  $V_{2\omega}$  signal versus  $V_{bg}$  for different applied heater currents ( $I_h$ ), at 40K for MATBLG and TDBLG device, respectively. (Bottom panels)  $V_{2\omega}$  signal normalized by the square of respective heater currents ( $I_h^2$ ) versus  $V_{bg}$ , both showing linear regime behavior.



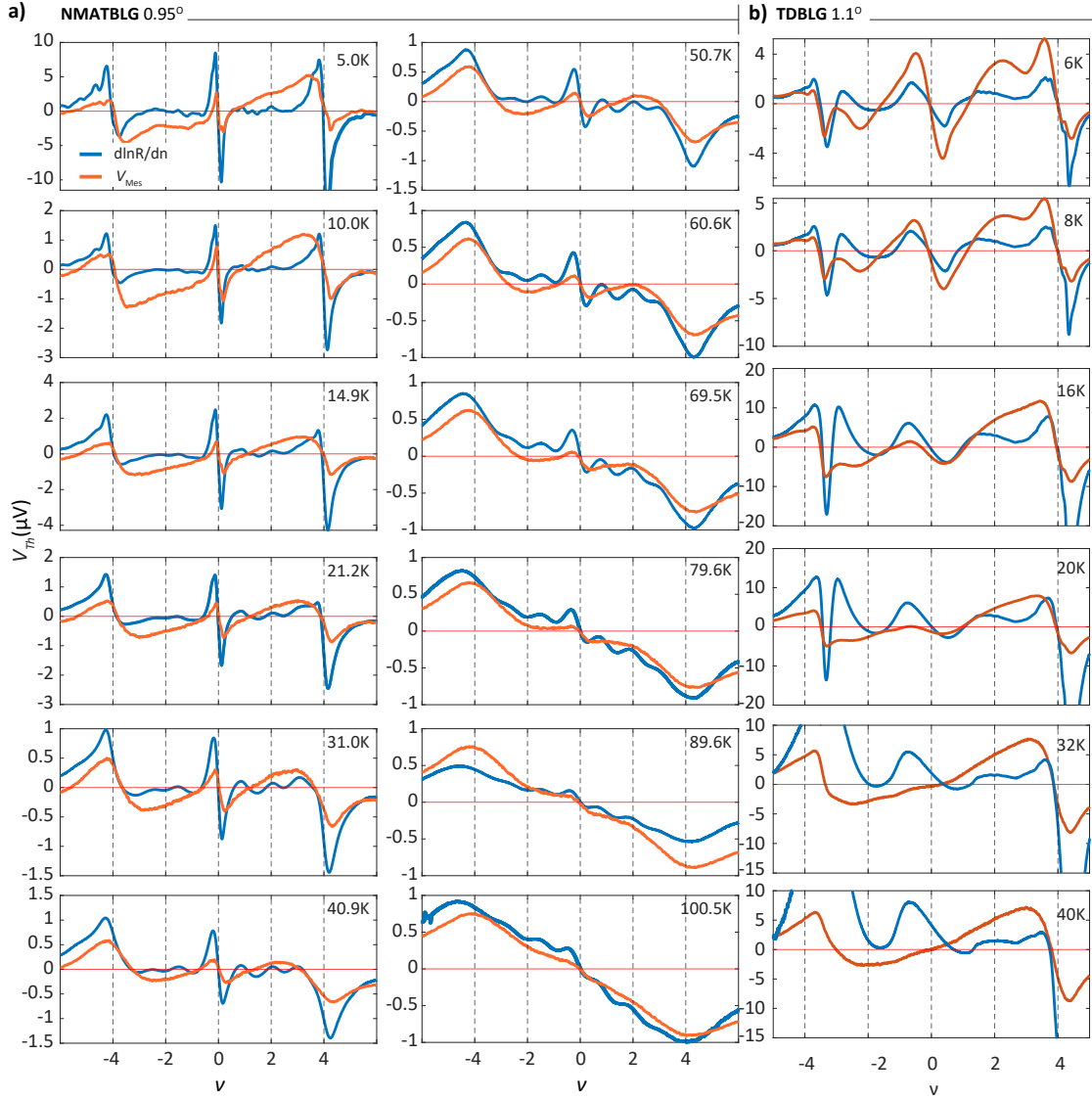
**Extended Data Fig. 3: Resistance ( $R$ ) and thermoelectric voltage ( $V_{Th}$ ) response for near MATBLG and TDBLG device: (a),(b),  $R$  and  $V_{Th}$  response of near MATBLG device, as a function of  $\nu$  at different temperatures, depicted in a shifted plot. The horizontal lines represent the zero reference at each temperature.  $R$  remains featureless at integer fillings, except for the Dirac point and  $\nu = \pm 4$ . The  $V_{Th}$  closely resembles that of MATBLG, exhibiting similar crossing points near  $\nu \approx \pm 1$ . However, in comparison to MATBLG, these crossing points near  $\nu \approx \pm 1$  show some variation with  $T$  and persist only up to 50K. Beyond 100K,  $V_{Th}$  as well as  $R$  begins to resemble a graphene-like spectrum. (c),(d),  $R$  and  $V_{Th}$  response of TDBLG device, as a function of  $\nu$  at different temperatures. In contrast to MATBLG or 0.95° near MATBLG, the additional crossing points in the band vanish at a much lower temperature, around 20K. Distinctively, above 20K,  $V_{Th}$  remain completely positive (negative) in the conduction (valence) band, which is opposite to how  $V_{Th}$  behaves for MATBLG and near MATBLG devices, but resembles semi-classical thermopower  $S_{SC}$  behaviour shown in Extended Data Fig. 4e.**



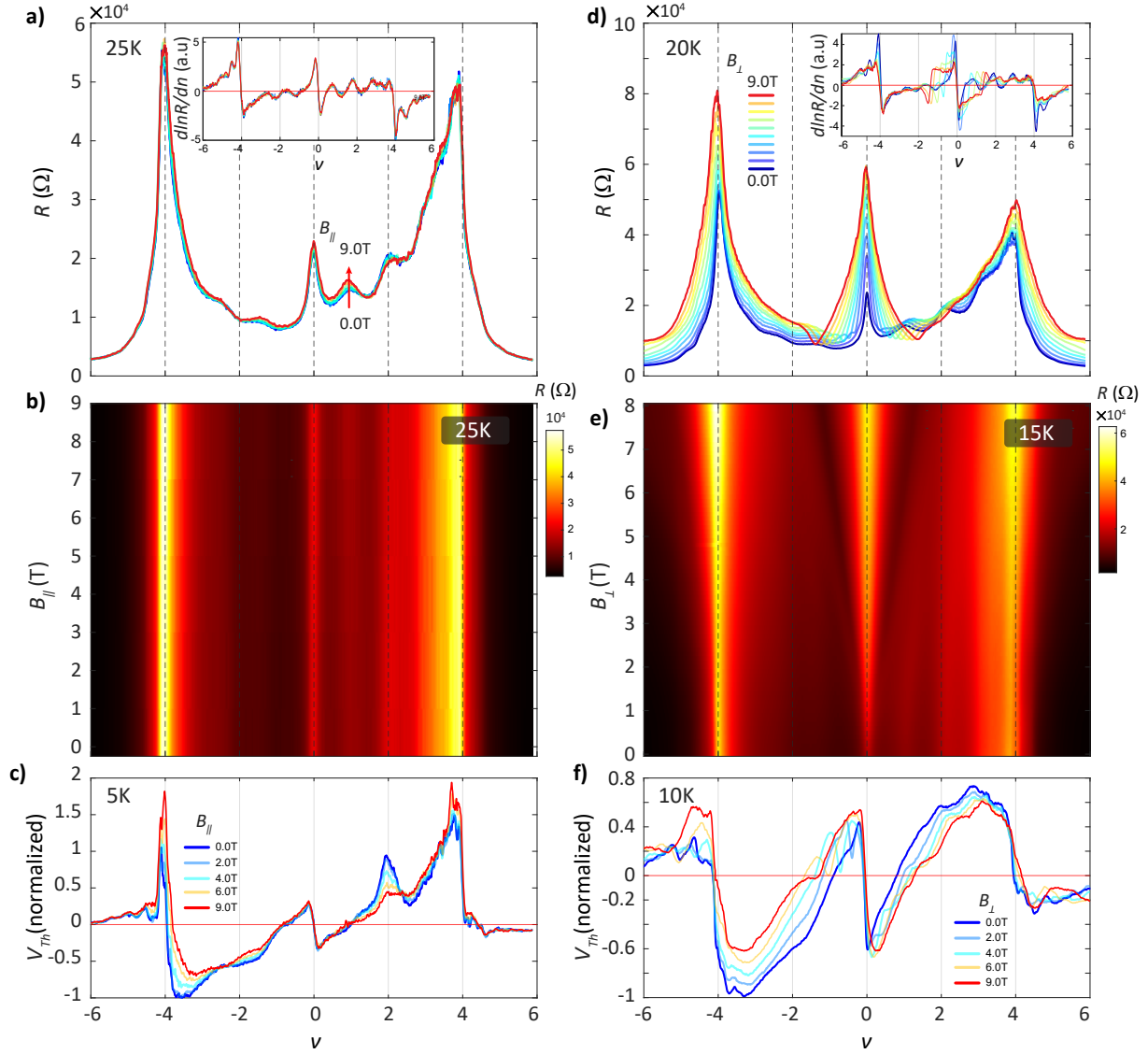
**Extended Data Fig. 4: Theoretical calculations of semiclassical thermopower :** (a) Continuum model DOS (by Zondiner, U. et al.) as a function of filling ( $\nu$ ) corresponding to twisted bilayer with twist-angle  $1.05^\circ$ . (b) Self-consistently solved chemical potential ( $\mu$ ) as a function of  $\nu$  at different temperatures. (c) and (d), respectively, show the numerator and denominator of Eq.1 in method section as a function of  $\nu$  at different temperatures. (e) Semi-classical thermopower with  $\nu$  at different temperatures plotted with an offset of 1 unit. The dashed horizontal lines correspond to the zero line references for respective temperatures.



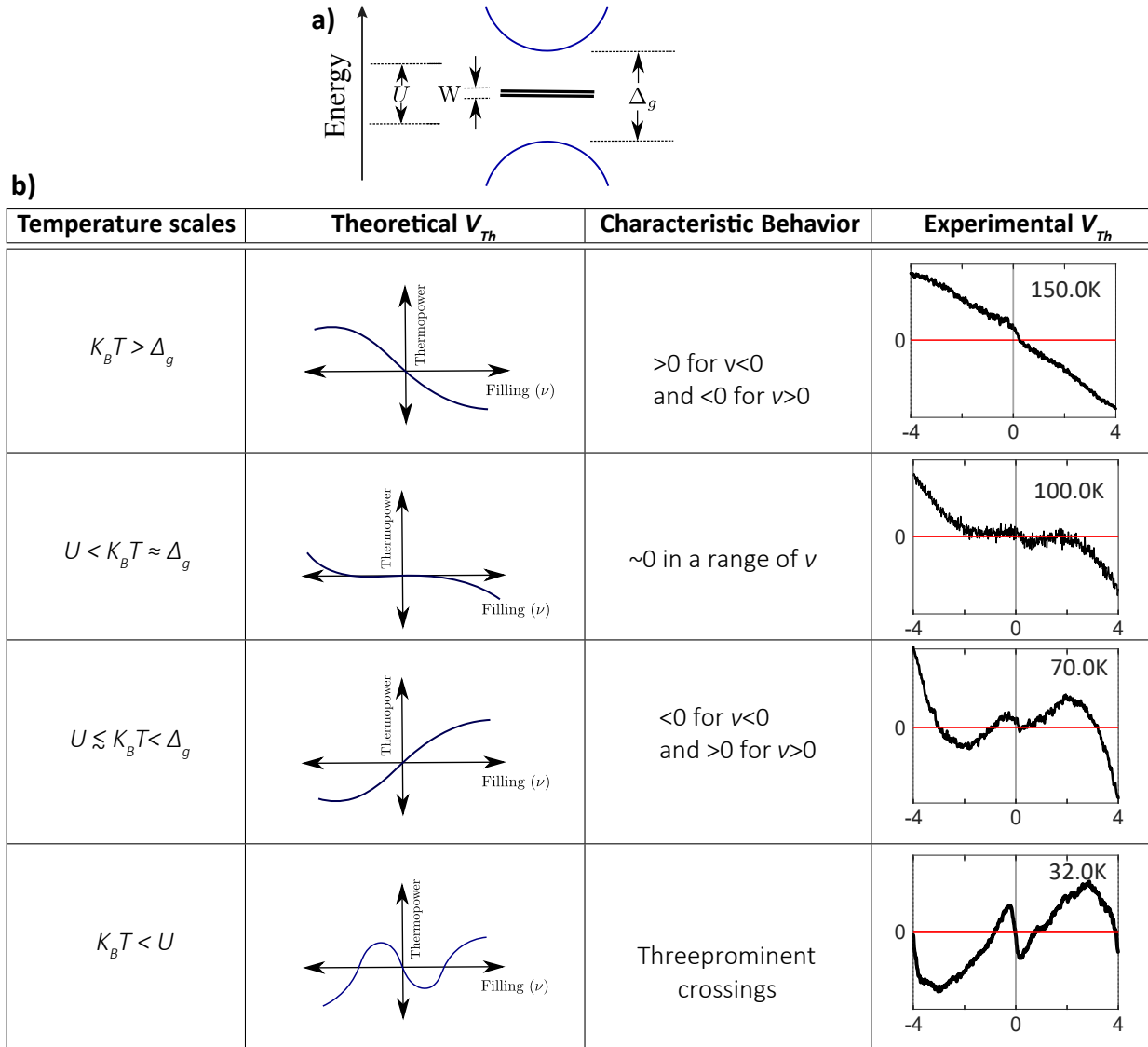
**Extended Data Fig. 5: Comparison between  $d\ln(R)/d\nu$  and measured  $V_{Th}$  for MATBLG device:** Comparison between measured  $V_{Th}$  signal and  $d\ln(R)/d\nu$  (both scaled to arbitrary units for visual clarity) at different temperatures. Blue and orange lines present the  $d\ln(R)/d\nu$  and  $V_{Th}$ , respectively. Below  $T < 120K$ , the measured thermopower (in orange solid line) lacks many of the crossing points predicted by  $d\ln(R)/d\nu$  (in blue solid line). Above  $120K$  the  $V_{Th}$  matches well with Mott relation with a graphene-like thermopower spectrum.



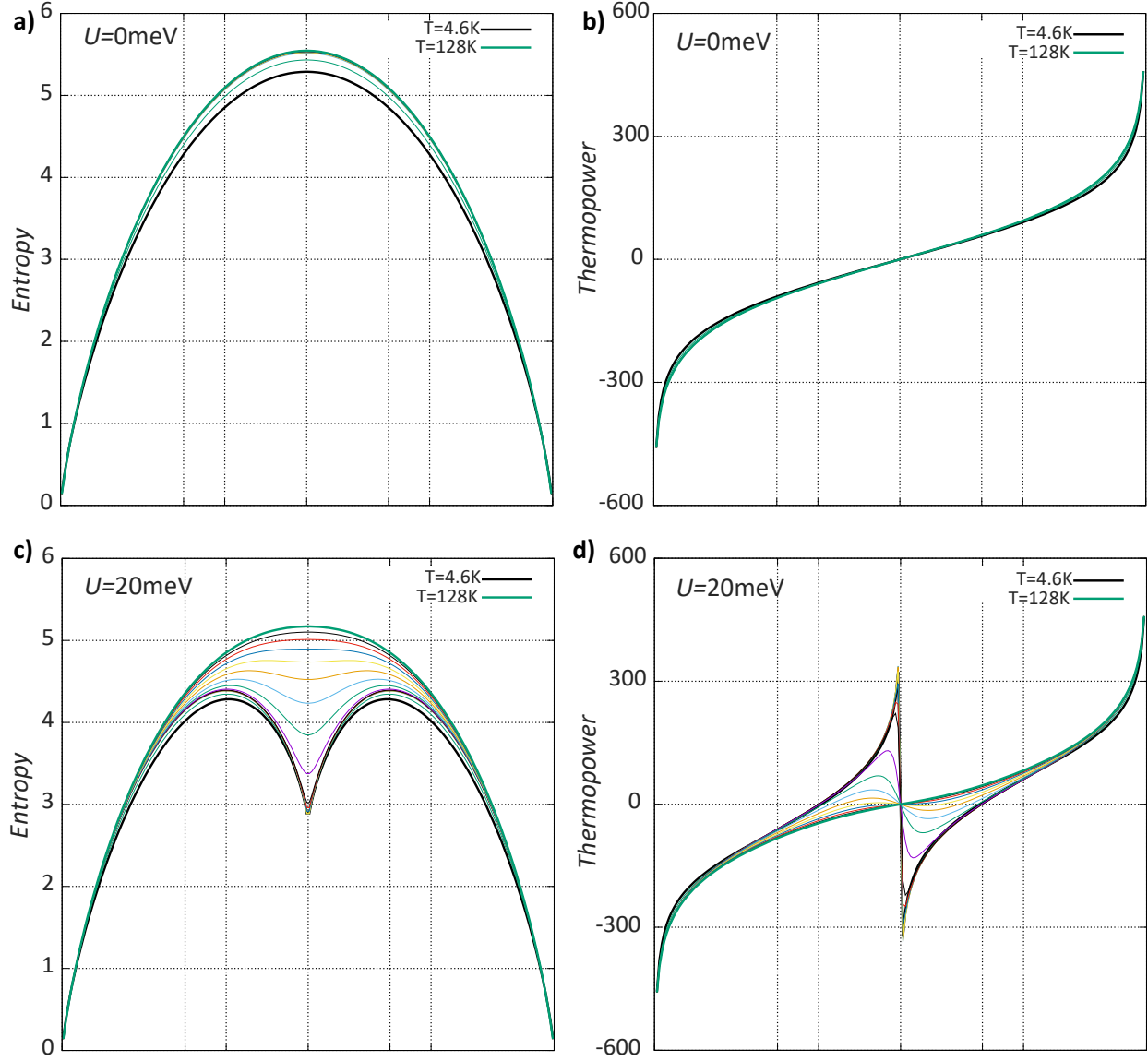
**Extended Data Fig. 6: Comparison between  $d\ln(R)/d\nu$  and measured  $V_{Th}$  for near MATBLG and TDBLG devices:** (a), (b), Comparison between measured  $V_{Th}$  signal and  $d\ln(R)/d\nu$  (both scaled to arbitrary units for visual clarity) at different temperatures for near MATBLG and TDBLG device, respectively. Blue and orange lines present the  $d\ln(R)/d\nu$  and  $V_{Th}$ , respectively. Mott violation can also be observed in near MATBLG devices. Below 50K,  $V_{Th}$  is generally more symmetric between the valence and conduction band, showing only one crossing point for each conduction and valence band, whereas,  $d\ln(R)/d\nu$  shows many crossing points. Here also, the measured  $V_{Th}$  matches quite well with  $d\ln(R)/d\nu$  at higher temperatures ( $> 90K$ ). In TDBLG, there is a distinct departure of the  $V_{Th}$  behaviour as compared to the MATBLG and near-MATBLG devices. At low temperatures ( $< 20K$ ),  $V_{Th}$  shows qualitative agreement with  $d\ln(R)/d\nu$  in terms of general peak and the number of zero crossing points (in the conduction band). At larger temperatures, the asymmetry in  $R$  (see Extended Data Fig. 3c) also manifests in the  $d\ln(R)/d\nu$  data, and  $d\ln(R)/d\nu$  mostly maintains a positive value over the whole density range of the flat band.



**Extended Data Fig. 7: Resistance and thermoelectric voltage with in-plane and out-of-plane magnetic fields:** (a) Resistance as a function of filling ( $\nu$ ) at different in-plane magnetic fields ( $B_{||}$ ) at 25K. Inset shows the  $d\ln(R)/d\nu$  at corresponding  $B_{||}$ . (b) 2D plot of Resistance as a function of  $\nu$  and in-plane magnetic field at 25K. Clearly, no noticeable change can be observed in the resistance or  $d\ln(R)/d\nu$  response, which starkly contrasts the  $V_{Th}$  response (see Figure 4 of the manuscript) with  $B_{||}$ . (c) Normalized  $V_{Th}$  with  $\nu$  for different  $B_{||}$  at 5K. Here also, the significant reduction of  $V_{Th}$  is clearly visible with  $B_{||}$ . (d) Resistance as a function of  $\nu$  at different out-of-plane magnetic fields at 20K. Inset shows the  $d\ln(R)/d\nu$  at corresponding  $B_{\perp}$ . (e) 2D plot of Resistance as a function of  $\nu$  and out-of-plane magnetic field at 15K. Except around the Dirac point, the signature of Landau levels can hardly be seen. The Mott formula, i.e.,  $d\ln(R)/d\nu$ , also fails to capture any decrement with  $B_{\perp}$  seen in Figure 4 in the main manuscript. (f) Evolution of normalized  $V_{Th}$  with  $B_{\perp}$  at 10K. Reduction of  $\sim 40\%$  in  $V_{Th}$  at  $\nu \simeq -3$  is apparent.



**Extended Data Fig. 8: Comparison between the measured  $V_{Th}$  and expected theoretical thermopower at different temperature regime:** (a) The key energy scales are bandwidth ( $W$ ) of the flatband, the interaction strength ( $U$ ), and the energy gap between the dispersing lower and upper bands ( $\Delta_g$ ). (b) (i) For  $k_B T > \Delta_g$ ; the high-temperature thermopower is positive for  $\nu < 0$  and negative for  $\nu > 0$  (as depicted in the 1st row). A characteristic of graphene-based systems where electron-hole symmetry renders one crossing at  $\nu = 0$  (Dirac point). (ii) For  $U < k_B T \approx \Delta_g$ ; the effect of flat bands starts contributing together with higher dispersing bands. However, both contributions have opposite signs, resulting in almost flat, close to zero  $V_{Th}$  (as shown in the second row). (iii) For  $U \approx k_B T < \Delta_g$ ; in this regime, the effect of the flatbands dominates over the higher energy bands. This leads to entropy being maximum at  $\nu = 0$  and single thermopower crossing at  $\nu = 0$  with positive for  $\nu > 0$  and negative for  $\nu < 0$  (as depicted in the third row). (iv)  $U < k_B T$ ; the interaction dominates and dictates the entropy, which results in three prominent crossings (as shown in the last row), and discussed in Extended data Fig. 9.



**Extended Data Fig. 9:** (a), (b) Behavior of entropy and thermopower for the four orbital atomic limit, where the onsite energies are  $-\epsilon_1 = -\epsilon_2 = \epsilon_3 = \epsilon_4 = 0.2\text{meV}$ , and Hubbard interaction  $U = 0\text{meV}$ . Different curves correspond to different temperatures: 4.6K, 7K, 14K, 16K, 19K, 21K, 23K, 26K, 35K, 46K, 58K, 70K, 81K, 93K, 104K, 116K and 128K. Thermopower has a zero crossing at only  $\nu = 0$ . (c),(d) Behavior of entropy and thermopower for the four orbital atomic limit where the onsite energies are  $-\epsilon_1 = -\epsilon_2 = \epsilon_3 = \epsilon_4 = 0.2\text{meV}$ , Hubbard interaction  $U = 20\text{meV}$ . Different curves correspond to different temperatures: 4.6K, 7K, 14K, 16K, 19K, 21K, 23K, 26K, 35K, 46K, 58K, 70K, 81K, 93K, 104K, 116K and 128K. Thermopower has a zero crossing at  $\nu = 0$  and  $\pm \frac{4}{3}$ .