Supporting Information: Anomalous electrical transport in orientationally controlled trinary hybrids of graphene and twisted bilayer Molybdenum disulphide.

Shaili Sett[1*], Sudipta Kundu[1], Saloni Kakkar[1], Navkiranjot Kaur Gill[1], Manish Jain[1] and Arindam Ghosh[1,2]

[1]Department of Physics, C. V. Raman Avenue, Indian Institute of Science, Bangalore-560012, India.

[2]Centre for Nano Science and Engineering, Indian Institute of Science, Bangalore-560012, India.

*Email: shailisett@iisc.ac.in

Section I: Material characterization and device fabrication

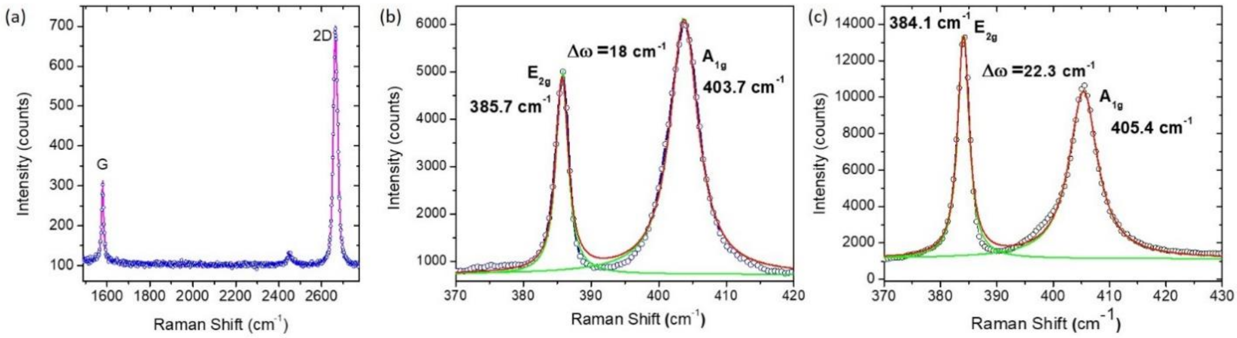

Figure S1. Raman Spectroscopy on 2D materials. (a) Monolayer graphene (b) monolayer MoS_2 and (c) bilayer MoS_2.

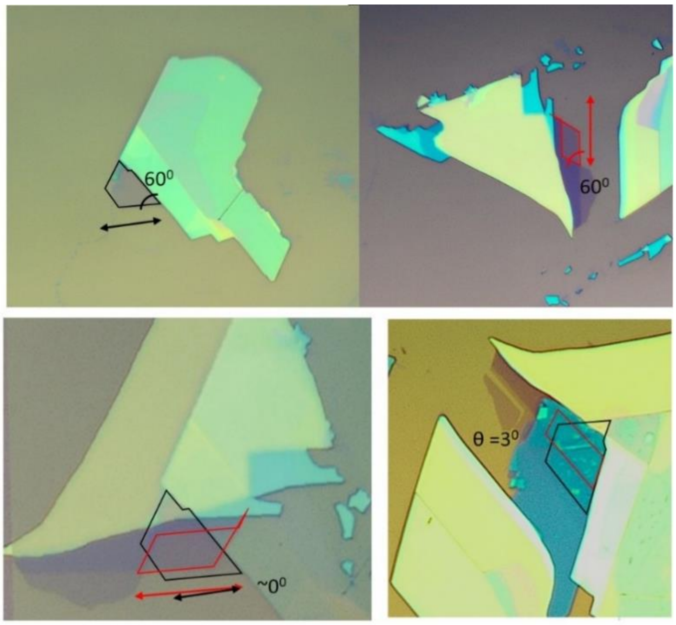

Figure S2. Transfer of twisted bilayer MoS_2 near 0^0. (a) and (b) Shows the monolayer flakes with the zig-zag edges marked with arrows. (c) Shows the overlaid images of the individual flakes with a near 0^0 twist angle with parallel alignment of the zigzag edges. (d) End product of Twisted bilayer MoS_2 after the transfer process. It shows the angle between the zig-zag edges of the two flakes to be 3^0.

Section II:

A: Electronic transport properties of the three hybrid devices.

Properties at 300 K	**Twist angle 58.5±0.5^0**	**Twist angle 3^0**	**Natural bilayer**
μ_e (cm^2/Vs)	3125	1180	1030
μ_h(cm^2/Vs)	290	635	625
σ (S) at Dirac point	7.7 x10^{-4}	7.8 x10^{-4}	1.55 x10^{-3}
Properties at 14 K	**Twist angle 58.5±0.5^0**	**Twist angle 3^0**	**Natural bilayer**
μ_e (cm^2/Vs)	7875	2144	2094
σ (S) at Dirac point	2.97 x10^{-4}	8.17 x10^{-4}	1.3 x10^{-3}

B: Estimating the magnitude of fluctuations.

The noise magnitude from V_g dependence is obtained by:

(i) The data is fitted with a smooth polynomial of third order.

(ii) The variance of the residual of the fitted curve is calculated. The variance gives the value of $\langle \delta R^2 \rangle$.

(iii) The mean value of the fitted curve corresponds to $\langle R \rangle$.

(iv) The magnitude of $\langle \delta G^2 \rangle$ is calculated using the relation, $\langle \delta G^2 \rangle = \langle \delta R^2 \rangle \, \langle R \rangle^4$

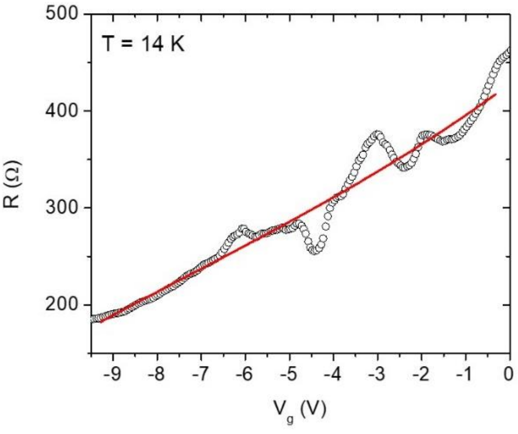

Figure S3. $R - V_g$ curve acquired over a small gate voltage window of 9 V for estimating the magnitude of fluctuations. The solid red line is a third order polynomial fit to the experimental data given in circles.

C: Reproducibility of the $R - (V_g - V_D)$ data.

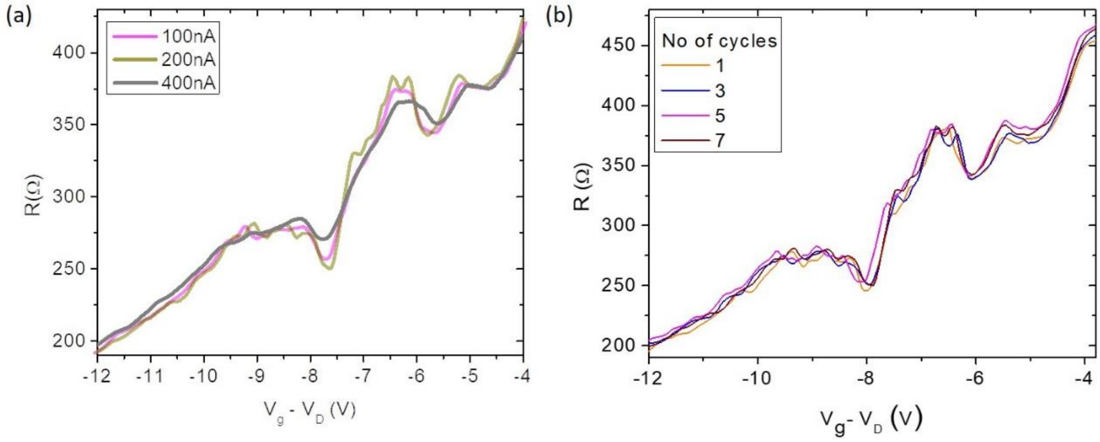

Figure S4. Magnified view of the corrugated regions in the $R - (V_g - V_D)$ curve at (a) different source currents and (b) for multiple cycles of data at 10 K.

Section III. Determination of the conduction band edge in twisted bilayer MoS_2.

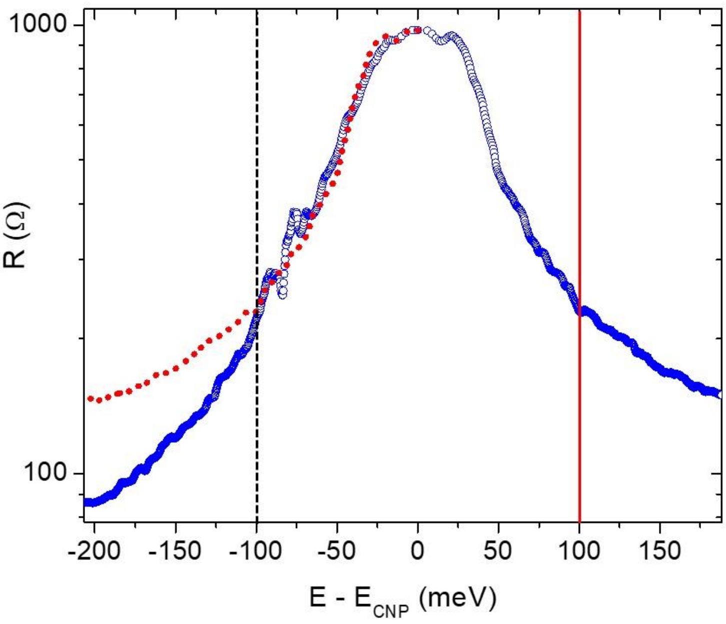

Figure S5. The $R-E$ curve of tbl_n60° device at 10 K. The blue symbol curve is the experimental data. The red dotted line is the mirror image of the electron doped $R-E$ characteristics which helps us to identify the electron-hole asymmetry arising due to screening by MoS_2. It occurs when the chemical potential of graphene aligns with the conduction band edge of MoS_2. The dotted line shows the position of asymmetry. The red line indicates the chemical potential of graphene when it enters the conduction band of MoS_2. This is the band offset (Δ) between graphene-MoS_2 heterostructure and it is ~ 100 meV.

Section IV: Estimation of effective gate capacitance in Gr-MoS_2 hybrid

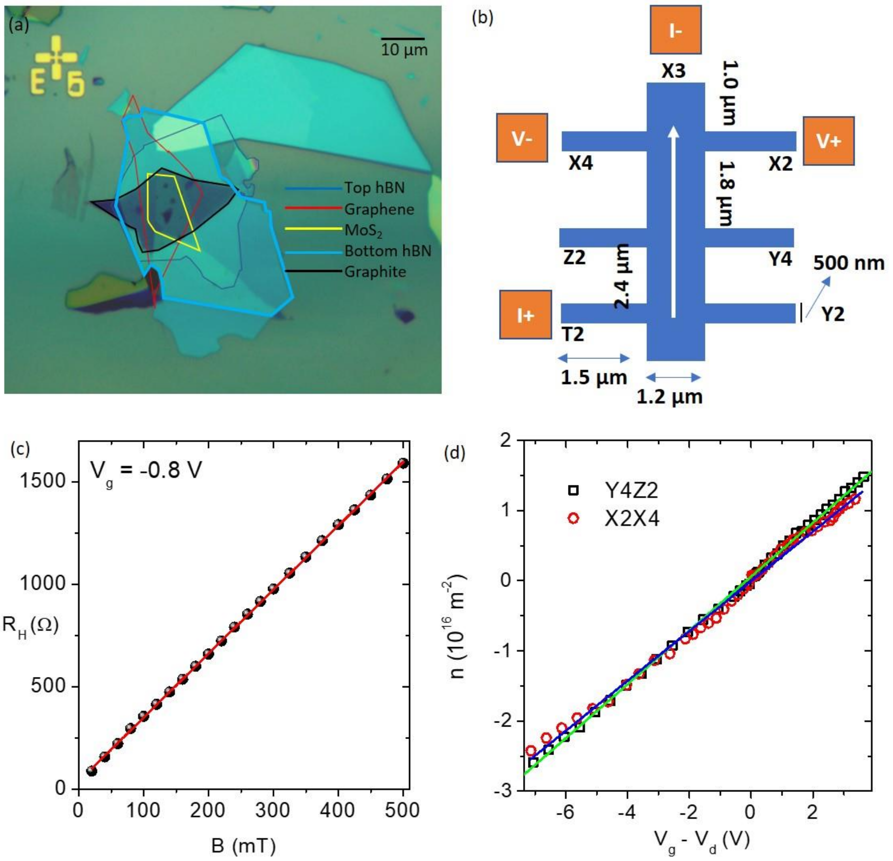

Figure S6. Hall effect measurements in a Gr-MoS_2 hybrid. (a) Optical image of the heterostructure. The 2D layers are arranged as follows: hBN-Gr-MoS_2-hBN-Graphite. (b) Schematic of Hall bar with electrodes labelled and device dimensions. (c) Hall resistance as a function of magnetic field at V_g = -0.8 V. (d) Number density (n) versus ($V_g - V_d$) plot for two different transverse channels, X4-X2 and Y4-Z2 as shown in schematic. The solid lines are fit to the data using equation 1.

<u>Determination of effective gate capacitance in Gr-MoS_2 hybrid:</u>

We use a hall geometry to determine the effective gate capacitance in the Gr-MoS_2 hybrid. We have used monolayer of graphene, monolayer MoS_2 and bottom hBN of thickness 35 nm (determined from atomic force microscopy). We use a local back gate with dielectric hBN (35 nm) with graphite contact. We carry out the measurements at low temperature of 15 K in the following way.

1) Measure Hall voltage as a function of magnetic field and calculate the Hall resistance. Using the equation, $R_H = 1/ne$, we find the number density n.

2) We determine n at different gate voltages (V_g) and plot n as a function of $V_g - V_d$. It follows the relation,

$$n = (V_g - V_d)C_{eff}/e \quad (1)$$

3) The slope of the straight line is equal to C_{eff}/e, where C_{eff} is the effective capacitance per unit area given by, $C_{eff} = \varepsilon_o \varepsilon_r / d_{eff}$, where d_{eff} is the effective width of the dielectric.

4) The value of d_{eff} obtained from the fit is 64 ± 2 nm while the hBN thickness used was 35 nm.

5) The value of d_{eff} obtained from the Hall experiment shows that the screening effects of MoS_2 influences the net gate capacitance experienced by the graphene layer.

Section V: Estimation of conduction band edge in twisted bilayer MoS_2 at 58.5°.

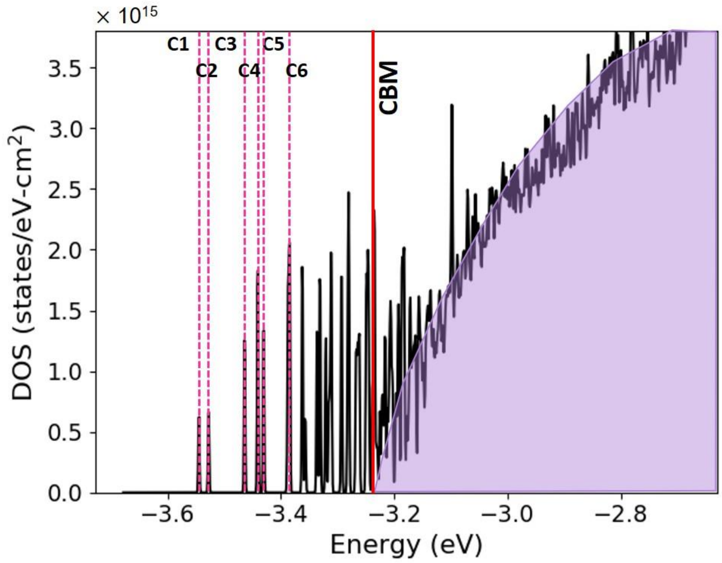

Figure S7. Density of states plot of twisted bilayer MoS_2 at 58.5°. We consider a parabolic DOS as in the conduction band which is shown by the shaded region. The conduction band minima/edge is marked as CBM in red at -3.241 eV. It is not exactly possible to identify the edge from where the continuum states emerge and this is a qualitative approach for calculating the position of the flatbands.