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Charge transport in ion-gated mono-, bi-, and trilayer MoS₂ field effect transistors

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Charge transport in MoS₂ in the low carrier density regime is dominated by trap states and band edge disorder. The intrinsic transport properties of MoS₂ emerge in the high density regime where conduction occurs via extended states. Here, we investigate the transport properties of mechanically exfoliated mono-, bi-, and trilayer MoS₂ sheets over a wide range of carrier densities realized by a combination of ion gel top gate and SiO₂ back gate, which allows us to achieve high charge carrier (>10¹³ cm⁻²) densities. We discuss the gating properties of the devices as a function of layer thickness and demonstrate resistivities as low as 1 kΩ for monolayer and 420 Ω for bilayer devices at 10 K. We show that from the capacitive coupling of the two gates, quantum capacitance can be roughly estimated to be on the order of 1 μF/cm² for all devices studied. The temperature dependence of the carrier mobility in the high density regime indicates that short-range scatterers limit charge transport at low temperatures.

Two-dimensional (2D) crystals of layered transition metal dichalcogenides (TMD) have gained significant interest due to their unique physical properties^{1,2}. Molybdenum disulfide (MoS₂), a semiconducting compound traditionally known for its lubricating properties, has been intensively investigated in its 2D crystalline form in the past few years. Field effect transistors of monolayer MoS₂ have shown remarkable characteristics with low off-current due to excellent gate electrostatics³. Recent studies on the implementation of MoS₂ and other semiconducting TMDs into integrated circuits⁴⁻⁷ and optoelectronic devices⁸⁻¹⁰ highlight their potential in future applications.

Electrostatically controlled switching and charge transport in mono- and few-layer MoS₂ devices are strongly influenced by various effects such as metal contacts^{11,12}, interface traps¹³⁻¹⁵, charged impurities^{3,13,15,16}, dielectric environment^{3,4,6,17,18}, and structural defects^{15,19-23} in the material. The contribution of these effects varies with the gate bias in a complex manner. At low gate biases near the sub-threshold region, the device resistance is strongly dominated by Schottky barriers at the metal contacts¹¹, band edge disorder and mid-gap states^{13,21,23-25}, which originate from structural defects, charge traps at the MoS₂/SiO₂ interface, and surface adsorbates^{13,14}. At higher charge densities where the channel conductivity exceeds e^2/h , conduction occurs via band-like carrier transport^{18,21,26,27}. In this regime, charged impurities, defects, and surface-optical phonons²⁸ limit charge carrier mobility in 2D MoS₂. All recent studies show that low temperature mobility of mono- and bilayer MoS₂ falls substantially below theoretically predicted values^{29,30}, suggesting that there is significant room for improvement in the device performance.

The use of high-κ dielectrics, such as HfO₂^{3,18,31} and Al₂O₃^{17,32}, as an encapsulating layer and gate barrier has been reported to be effective in enhancing carrier mobility, potentially due to dielectric screening of charged impurities and suppression of homopolar optical phonon modes^{18,33}. The high-κ gate dielectric also allows access to the high density regime where transport is band-like and the intrinsic properties of the material can be investigated. In order to achieve even higher charge densities, ionic gating has been used in different 2D materials³⁴⁻³⁹. Previous studies using polymer electrolyte⁴⁰, ionic liquid^{27,38,41,42}, and ion gels⁴³ have demonstrated effective switching and high doping levels of up to ~10¹⁴ cm⁻² in few-layer MoS₂ devices^{42,43} to realize flexible transistors⁴³, stable p-n junctions^{35,41,44}, light emitting devices^{32,35,44}, and superconductivity²⁷.

In this article, we report on the low temperature transport characteristics of mono-, bi-, and trilayer MoS₂ devices in the high doping regime using ion gel gating. We show that large capacitive coupling of the ion gel in

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conjunction with additional electrostatic control by the back gate allows systematic investigation of charge transport over a wide range of carrier densities. Resistivities as low as 1 k Ω and 420 Ω are realized in highly doped mono- and bilayer MoS₂ at low temperature, respectively. From the capacitive coupling of the two gates, we estimate the quantum capacitance, which is a measure of the density of states in these materials. Further analysis of the temperature- and density-dependent field effect mobility reveals that short-range scatterers severely limit carrier mobility at low temperatures in all these devices.

Results

The thickness of the exfoliated flakes is estimated by optical contrast and then further confirmed by the peak separation between the A_{1g} and E_{2g}¹ peaks in the Raman spectrum⁴⁵. In agreement with previous studies¹³, the peak separation was found to be 18.3 cm⁻¹, 21.2 cm⁻¹, and 23.1 cm⁻¹ for monolayer (1 L), bilayer (2 L), and trilayer (3 L) samples, respectively. The optical images of the samples covered with a thin layer of ion gel are shown in figure 1a (see methods section for details). Figure 1b shows the transfer curves of mono-, bi-, and trilayer devices as a function of the top gate voltage (V_{tg}) applied to the ionic gate at room temperature. For bilayer and trilayer devices, both electron and hole branches are observed at positive and negative gate voltages, respectively, demonstrating the wide tunability of the chemical potential achieved by ion gel. The monolayer device only shows the electron branch in the top gate bias window studied due to its large band gap.

Assuming that the variation of the electrostatic potential is negligible, the bandgap of bilayer and trilayer samples can be roughly estimated to be 1.63 eV and 1.32 eV from the threshold voltage difference for the electron and hole branches of the transfer curve (Supplementary Information, Figure S3)^{35,39,41}. These values are consistent with the optical gap previously measured by photoluminescence spectroscopy⁴⁶. Figure 1d displays the temperature dependent transconductance of a bilayer device, obtained by sweeping the top gate voltage (4.5 mV/s) at a fixed temperature. Below $T \sim 210$ K, the top gate modulation becomes negligible, suggesting the freezing of the ions in the gel matrix. This immobilization of ions allows us to achieve stable high doping in the channel and fine control of the carrier density by the back gate below this temperature.

The dual gating behavior of the devices was studied by cooling them below the critical freezing point at a fixed top gate voltage and measuring the transfer characteristics by back gate sweeps at various temperatures. To study the device over a wide range of carrier densities, the sample was brought to room temperature and the top gate voltage was set to a desired value and held until the displacement current was negligible. The sample was then cooled below the freezing point of the ion gel for measurements. The above procedure was repeated with different top gate biases. Figures 2a–c show the back gate transfer characteristics of the devices at different top gate biases. At negative V_{tg} , transitions from the insulating to the conducting state can be observed with on-off ratios of $>10^5$ for all devices. The off-state current was limited by the gate leakage current, which was typically below 100 pA at temperatures below 200 K (See Supplementary Figure S7). It is worth noting that all samples exhibit

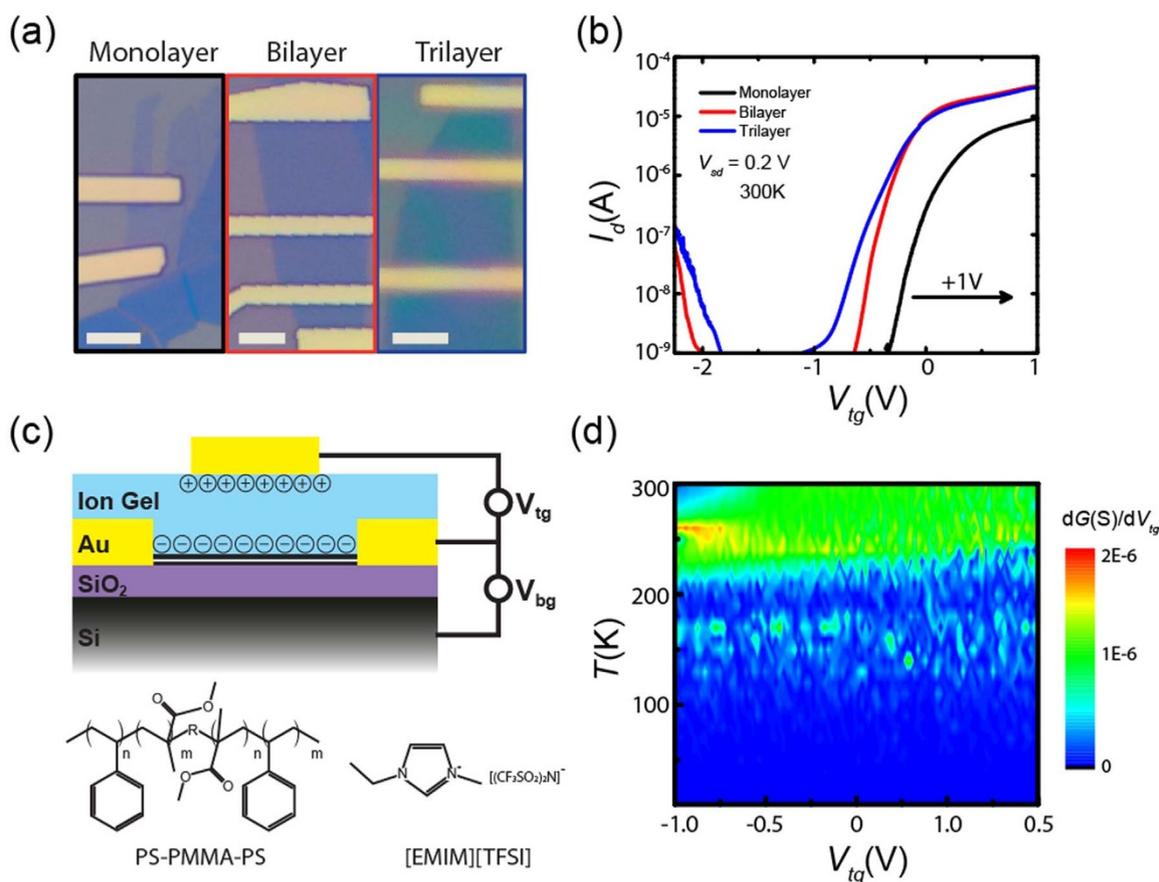


Figure 1 | Ionic gating of MoS₂ devices. (a) Optical images of mono-, bi-, and trilayer MoS₂ transistors covered with ion gel. The scale bars is 5 μ m.

(b) Ambipolar field-effect characteristics at room temperature. The curve for the monolayer sample is shifted by 1 V to the left for clarity.

(c) Schematic illustration of a dual gated transistor and chemical structures of the copolymer and the ionic liquid used as the top gate.

(d) Transconductance of a bilayer device while sweeping V_{tg} at a fixed temperature. Below ~ 200 K, the ions are frozen out and top gate variations have no effect on the sample's conductance.

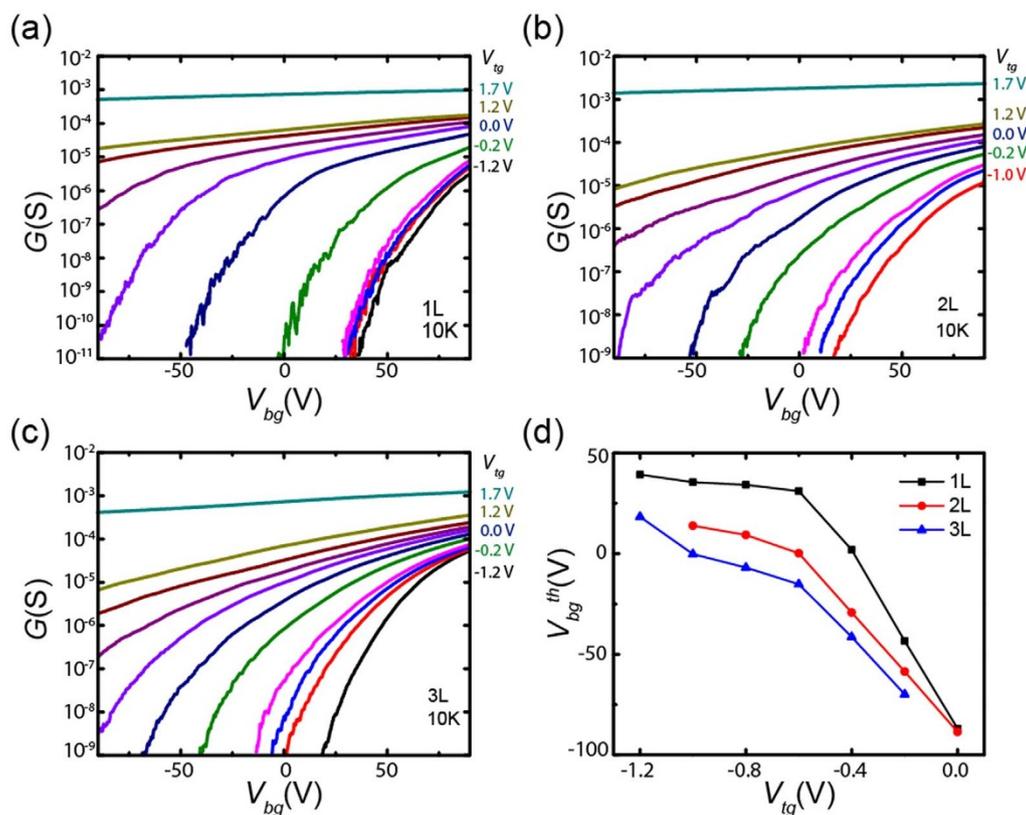


Figure 2 | Shift of threshold voltages. (a–c) Channel conductivity as a function of V_{bg} at different V_{tg} (–1.2 V, –1.0 V, ... 1.0 V, 1.2 V, 1.7 V) for 1–3 L MoS₂ devices at 10 K. (d) The threshold voltage obtained from the cutoff in (a–c) as a function of V_{tg} . For positive V_{tg} , V_{bg}^{th} cannot be extracted since the sample is always in the on-state.

low sheet resistivity (<1 k Ω) at large top gate bias and low temperature. These values are among the lowest reported for monolayer and bilayer MoS₂ so far.

The threshold back gate voltage V_{bg}^{th} , which we defined here to be the bias voltage required to achieve a critical device current of 100 pA, shifts towards negative gate bias with increasing top gate voltage as shown in Figure 2d. Note that for positive top gate voltages the devices remain in the conducting state for all back gate biases. It can also be seen that the shift of V_{bg}^{th} is non-monotonous with an apparent change in the slope at around $V_{tg} = -0.6$ V. We attribute this behavior to different capacitive coupling of the top and back gate with shift in E_F and the corresponding changes in the quantum capacitance³⁴ C_q of MoS₂. The total top gate capacitance between ion gel and MoS₂ channel C_{tot}^{top} can be described as a series connection of C_q , which is proportional to the density of states (DOS) of the material, and the geometric capacitance of the ion gel C_{ig} . Similarly, the back gate capacitance C_{tot}^{back} can be described as quantum capacitance and the oxide capacitance C_{ox} in series. Ignoring screening effects, the top and back gate capacitances are given by:

$$C_{tot}^{top} = \left(\frac{1}{C_q} + \frac{1}{C_{ig}} \right)^{-1}$$

$$C_{tot}^{back} = \left(\frac{1}{C_q} + \frac{1}{C_{ox}} \right)^{-1}$$

Here, we can consider two regimes depending on the relative magnitude of the capacitances. When the channel is depleted and E_F lies in the band gap, C_q is very small due to the low density of mid-gap states³⁹. This results in $C_q \ll C_{ig}$ and thus $C_{tot}^{top} \sim C_q$. Note that C_{tot}^{back} is affected by the same effect, but to a lesser extent because C_{ox} is

intrinsically small. As a result, the ratio between C_{tot}^{top} and C_{tot}^{back} remains low. In the other limit, when the sample is strongly doped and E_F is in the conduction band, C_q significantly increases such that C_{tot}^{top} has contributions from both C_q and C_{ig} while $C_{tot}^{back} \sim C_{ox}$. In this regime, the channel conductivity is much more efficiently tuned by the top gate bias as expected from the large capacitance of the ion gel. Thus, the ratio between C_{tot}^{top} and C_{tot}^{back} increases with increasing top gate voltage due to increase in C_q .

In the following, we analyse the results in the high doping regime. Figures 3a–c show the color-coded map of the channel conductivity as a function of top and back gate biases. As indicated by the black dashed lines along constant conductivity, the top gate is 50 to 100 times more efficient compared to the back gate. The ratios $\Delta V_{bg} / \Delta V_{tg}$, where ΔV_{bg} and ΔV_{tg} denote the corresponding gate voltage difference to achieve the same change in conductivity in the metallic regime, are obtained from the slopes of the dashed lines in Figure 3. This ratio is determined to be ~ 86 for monolayer, ~ 64 for bilayer and ~ 43 for trilayer device.

In order to show the full range of conductivity accessible by varying the two voltages, we offset the back gate transfer curves along the horizontal axis to highlight their linear (i.e. mobility saturation) behavior (Figure 4a and b). The insets show the transition from insulating to metallic conduction regimes where the temperature coefficient changes sign¹⁸. The crossover occurs at $V_{bg} \sim 80$ V and $V_{tg} = 0$ V for monolayer and $V_{bg} \sim 25$ V and $V_{tg} = 0$ V for bilayer. In accordance with the previous results^{18,26,27} the crossing points are at resistances on the order of h/e^2 . Above the crossover point, conductivity decreases with increasing temperature, indicating phonon limited, metal-like transport. At lower charge carrier densities, increasing conductivity with increasing temperature suggests thermally activated transport and conduction by variable range hopping^{13,21,23–25}. As can be seen from Figure 4b, ionic gating allows

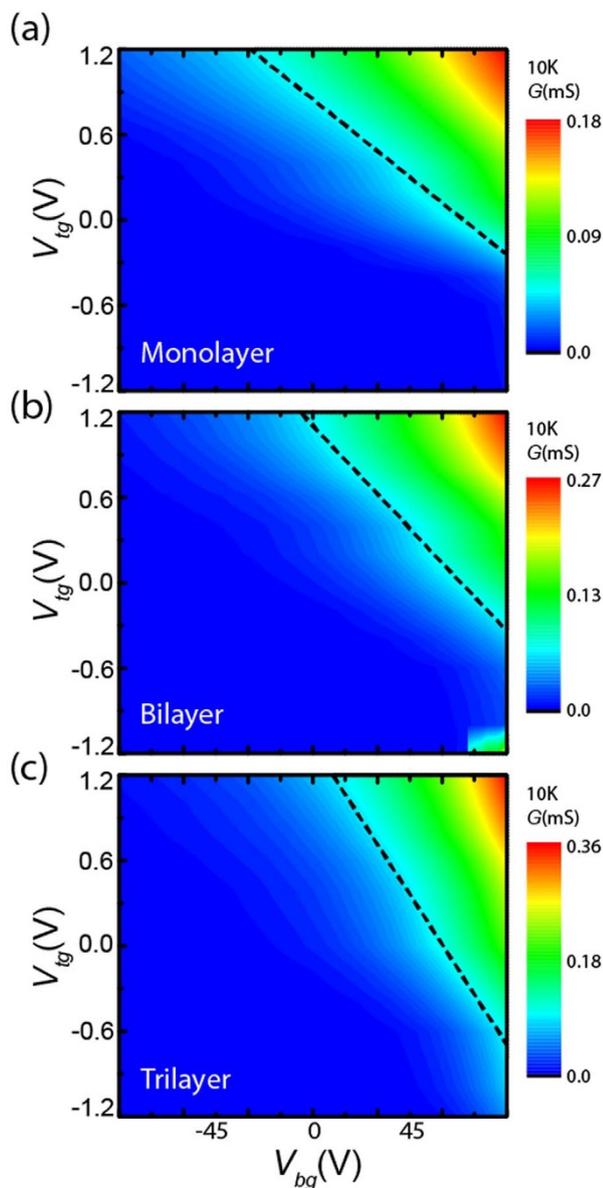


Figure 3 | Gate coupling for different number of layers. Color-coded map of the dual gate characteristics for (a) monolayer, (b) bilayer and (c) trilayer MoS₂. After setting the top gate voltage at room temperature and cooling the sample down to $T = 10$ K, the back gate bias was varied. The dashed black lines highlight constant conductivity just above the onset of conduction.

access to conduction regimes far beyond the crossover point where transport properties remains largely unexplored.

Field effect mobility and its dependence on carrier density and temperature offer insight into the fundamental transport properties of MoS₂⁴⁷. The gate bias and temperature dependence of the field effect mobility ($\mu_{FE} = (1/C_{ox}) * d\sigma/dV_{bg}$, where $C_{ox} = 11.5$ nF/cm² is the back gate capacitance and σ is the channel conductivity) for mono- and bilayer MoS₂ is shown in Figures 4c–f. The mobility initially increases, and eventually saturates to a constant value at with increasing voltages. The mobility saturation can also be seen as a function of temperature below 30 K (Figure 4e and f). Note that the mobility becomes independent on both carrier density and temperature in this regime. The saturation values are about 230 cm²/Vs, 450 cm²/Vs, and 820 cm²/Vs for 1, 2, and 3 L devices, respectively (see Supplementary Information for 3 L device data).

Discussion

The difference in the gate coupling ratio for 1–3 L devices can originate from various effects, such as the superposition of the two gate fields, changes of dielectric constants, different gate separation, and changes of C_{ig} or C_q as a function of layer thickness. Using a simple relationship $C_{tot}^{top} \Delta V_{ig} = C_{tot}^{back} \Delta V_{bg}$ from the capacitor model and assuming an ion gel capacitance⁴³ to be $C_{ig} = 10.7$ μ F/cm², we estimate the quantum capacitance of mono-, bi-, and trilayer MoS₂ to be 1.0 μ F/cm², 0.8 μ F/cm², and 0.5 μ F/cm², respectively. These are rough estimates but are on the same order of magnitude as the theoretical prediction⁴⁸. The trend contradicts the expectation that DOS increases with increasing thickness⁴⁹, suggesting that ionic gate is effective only for the top layer, which is in direct contact with the gel. Consequently, two parallel channels that are independently controlled may be formed in multilayers. More accurate measurement of the quantum capacitance requires precise measurement of C_{ig} , (see Supplementary Information for Hall measurement results).

The mobility saturation shown in Figure 4 agrees with the earlier studies and indicates that at low temperatures and in the high carrier density regime, charge transport in these devices is dominated by short-range scattering²¹. The short-range scatterers may be structural defects such as sulfur vacancies and charged impurities which are fully screened by the free carriers. Our results show this behavior not only in monolayer but also in bilayer and trilayer MoS₂ devices, which indicates that intrinsic scattering mechanisms limit the device performance at low temperatures. The increase in saturation mobility with increasing layer number suggests that short-range scatterers are primarily present at the outer surface of the flakes possibly due to exposure to the ambient and solvent. It should be noted that despite the high concentration of ions, which are effectively charged impurities at the surface of the MoS₂, the low temperature mobility remains nearly constant with top gate bias. This observation suggests that the contribution of long-range scatterers (e.g. charged impurities) at the interface is less dominant compared to short-range scatterers (i.e. structural defects and fully screened charged impurities in the material) in this regime^{18,28}. For temperatures above 100 K, the mobility decreases rapidly due to electron-phonon scattering, which can be described by a power law dependence $\mu \sim T^{-\gamma}$. We found that while the Y factor is nearly constant with back gate voltage (Figure 4e), it is strongly dependent on top gate voltage. In bilayer MoS₂, it increases from 1.94 to 2.85 with increasing V_{ig} from 1.65 to 3 V (Figure 4f), similar to the trends observed in previous studies^{18,21}.

In summary, we show that dual gating of mono- and few-layer MoS₂ using ion gel allows access to a wide range of carrier densities. We demonstrate low device resistivities of 1 k Ω and 420 Ω in mono- and bilayer devices at low temperatures, respectively. The analysis of the dual gating behavior of the devices reveals that the gate coupling is strongly dependent on the doping level and the number of layers. From the coupling ratios, we estimate the quantum capacitance to be on the order of 1 μ F/cm² for all devices. Our transport results further shed light on the factors limiting carrier mobility in these materials in the highly conducting regime.

Methods

Atomically thin flakes of MoS₂ sheets were mechanically exfoliated from bulk crystals (SPI supplies) and subsequently deposited onto silicon substrate with 300 nm thermal SiO₂. Pure gold contacts (50 nm) for source, drain, and side gate electrodes were fabricated using standard electron beam lithography and thermal evaporation. After lift-off, all devices were annealed in nitrogen atmosphere at 200°C for 2 hours. The ion gel solution was prepared by mixing the polymer PS-PMMA-PS and the ionic liquid EMIM-TFSI (Figure 1c) into an ethyl propionate solvent (weight ratio of polymer: ionic liquid: solvent = 0.7 : 9.3 : 90)¹⁵. A film of this solution was spin-coated onto the devices and dehydrated in nitrogen gas for an hour to remove molecular moisture. Transport measurements were performed in vacuum inside a Helium 4 cryostat with variable temperature insert.

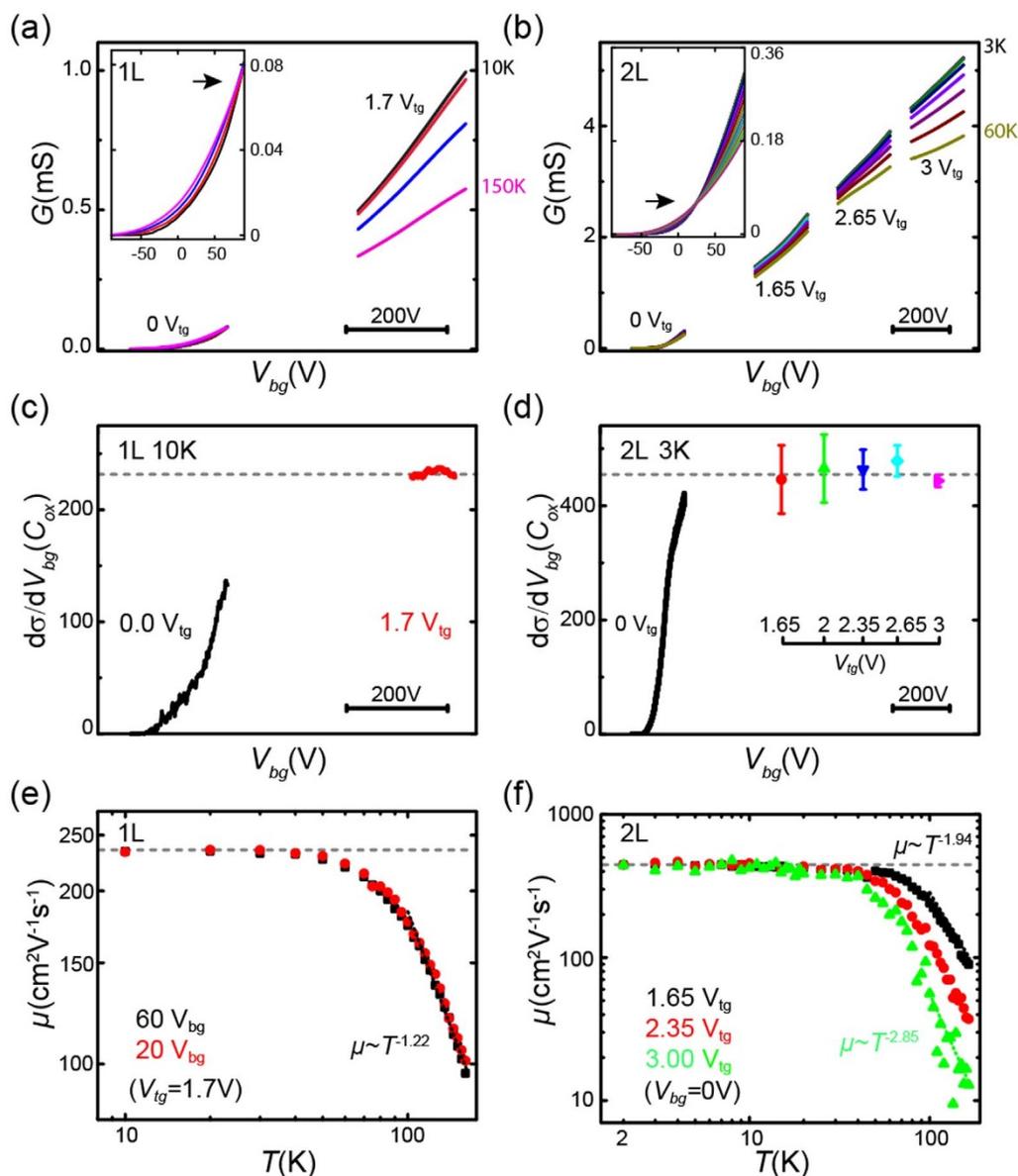


Figure 4 | Transport characteristics at high charge carrier densities. (a, b) Transfer curves of mono- and bilayer samples based on back gate sweeps at different top gate biases. The curves are displaced horizontally to highlight the linear regime where mobility saturates. The arrows in the insets of (a) and (b) indicate the metal-insulator transition point. (c, d) Differential conductivity in units of the oxide capacitance at low temperature as a function of back gate voltages at different top gate biases. Error bars are obtained from the variation of the mobility values as a function of the back gate voltages. (e, f) Temperature dependence of field effect mobility for mono- and bilayer devices. Power law fits $\mu \sim T^{-1.22}$ and $\mu \sim T^{-1.94}$ are shown to highlight the changes in the phonon damping factors.

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Author contributions

G.E. supervised the project, G.E. and H.S. designed the experiment, L.C. and S.W. prepared the samples, J.P. and T.T. supplied the ion gel and advised on the usage, L.C. and H.S. performed the measurements, G.E., L.C. and H.S. analysed the data. L.C., H.S., J.P., S.W., B.O., T.T. and G.E. discussed the results and contributed in writing the manuscript. All authors have given approval to the final version of the manuscript.

Additional information

Supplementary information accompanies this paper at <http://www.nature.com/scientificreports>

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