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

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Two-dimensional (2D) crystals of layered transition metal dichalcogenides (TMD) have gained significant interest due to their unique physical properties. 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 electrostatics. Recent studies on the implementation of MoS2 and other semiconducting TMDs into integrated circuits and optoelectronic devices 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, interface traps, charged impurities, dielectric environment, and structural defects 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 contacts, band edge disorder and mid-gap states, which originate from structural defects, charge traps at the MoS2/SiO2 interface, and surface adsorbates. At higher charge densities where the channel conductivity exceeds \( \epsilon'\hbar \), conduction occurs via band-like carrier transport. In this regime, charged impurities, defects, and surface-optical phonons 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, suggesting that there is significant room for improvement in the device performance.

The use of high-\( \kappa \) dielectrics, such as HfO2 and Al2O3, 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. Previous studies using polymer electrolyte, ionic liquid, and ion gels have demonstrated effective switching and high doping levels of up to \( \sim 10^{14} \text{ cm}^{-2} \) in few-layer MoS2 devices to realize flexible transistors, stable p-n junctions, light emitting devices, and superconductivity.

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
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 kΩ and 420 Ω are realized in highly doped mono- and bilayer MoS$_2$ 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 spectrum$^{45}$. In agreement with previous studies$^{13}$, 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 spectroscopy$^{46}$. 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...
low sheet resistivity (<1 kΩ) 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 capacitance $C_q$ 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 states. This results in $C_q \ll C_{ig}$ and thus $C_{tot}^{back} \sim C_q$. 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 $C_{tot}^{back}$.

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 $\Delta V_{bg}$ and $\Delta V_{tg}$ 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 $\sim 86$ for monolayer, $\sim 64$ for bilayer and $\sim 43$ 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 sign. 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, the crossing points are at resistances on the order of $\nu/e^2$. 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. As can be seen from Figure 4b, ionic gating allows

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

Field effect mobility and its dependence on carrier density and temperature offer insight into the fundamental transport properties of MoS₂. The gate bias and temperature dependence of the field effect mobility (\( \mu_{\text{FE}} \)) can be described by a power law dependence

\[ \mu_{\text{FE}} \propto \frac{1}{C_{\text{ox}}} \sigma \frac{d\phi}{dV_{\text{bg}}} \]

where \( C_{\text{ox}} \) is the back gate capacitance and \( \sigma \) is the channel conductivity. The mobility initially increases, and eventually saturates to a constant value at with increasing voltages. The mobility saturation can be seen as a function of temperature below 30 K (Figure 4e). 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 field effect mobility (\( \mu_{\text{FE}} \)) and temperature dependence of the field effect mobility (\( \mu_{\text{FE}} \)) can be described by a power law dependence

\[ \mu_{\text{FE}} \propto \frac{1}{C_{\text{ox}}} \sigma \frac{d\phi}{dV_{\text{bg}}} \]

where \( C_{\text{ox}} \) is the back gate capacitance and \( \sigma \) is the channel conductivity. The mobility initially increases, and eventually saturates to a constant value at with increasing voltages. The mobility saturation can 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).

**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 10:90). 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 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.

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