Anomalous Conductance near Percolative Metal–Insulator Transition in Monolayer MoS$_2$ at Low Voltage Regime

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Supporting Information

ABSTRACT: Conductivity of the insulating phase increases generally at an elevated drain–source voltage due to the field-enhanced hopping or heating effect. Meanwhile, a transport mechanism governed by percolation in a low compensated semiconductor gives rise to the reduced conductivity at a low-field regime. Here, in addition to this behavior, we report the anomalous conductivity behavior to transform from a percolative metallic to an insulating phase at the low voltage regime in monolayer molybdenum disulfide (MoS$_2$). Percolation transport at low source–drain voltage is governed by inhomogeneously distributed potential in strongly interacting monolayer MoS$_2$ with a substrate, distinct from the quantum phase transition in multilayer MoS$_2$. At a high source–drain voltage regime, the insulating phase is transformed further to a metallic phase, exhibiting multiphases of metallic–insulating–metallic transitions in monolayer MoS$_2$. These behaviors highlight MoS$_2$ as a model system to study various classical and quantum transports as well as metal–insulator transition in two-dimensional systems.

KEYWORDS: monolayer MoS$_2$, anomalous conductivity behavior, voltage-induced transitions, percolation, metal–insulator transition

Since the first application of field-effect transistors,† transition metal dichalcogenides (TMDs) have been established in extensive studies to have diverse exotic physical phenomena.‡–§ In particular, the existence of the stable form of monolayer having distinct properties from multilayers, e.g., direct band gap with a strong spin–orbit coupling, provides an attractive playground for the observation of exceptional phenomena such as valley-selective circular dichroism, piezoelectricity, 7 charged excitons, 8 etc. The charge transport in monolayers is strictly confined in two dimensions (2D) of atomic thickness. Most carriers are thus easily disturbed by a variation of external potential such as the substrate and material surface, resulting in not only lower mobility but also some anomalous transports, which can hardly be observed in multilayer systems.

The conductivity in the insulating phase, in general, increases with the driving excitation voltage (or field) due to the field-enhanced hopping or heating effect. The opposite behavior of decreasing conductivity with a field at the low-field regime observed in low compensated semiconductors is, thus, anomalous and is attributed to the existence of certain structures playing an important role in the hopping transport, called “dead ends” or “returns”, in a percolation picture. 9 Similar behavior is observed in our present monolayer molybdenum disulfide (MoS$_2$), more importantly, with an unusual transition from a metallic to an insulating phase driven by a field. This transition along with the anomalous conductivity behavior in the insulating phase can be also explained in the percolation picture. This feature has not been observed in multilayer MoS$_2$, and the metal–insulator transition (MIT) in multilayer systems is interpreted as a quantum phase transition (QPT) 10 in contrast to the present monolayer case.

The origin of the MIT phenomena in 2D has been disputed for several decades since the first experimental observation which showed possible MIT in Si metal-oxide field-effect transistors (Si-MOSFETs). 11 This observation was the
contrary result from the renowned scaling theory of localization$^{12}$ predicting the absence of MIT in 2D and consequently attracted a great deal of immediate interests in the underlying mechanism of the phenomenon. Several scenarios have been suggested, including QPT,$^{11,13}$ percolation,$^{14,15}$ crossover at finite temperature,$^{16}$ etc. Although extensive data have been accumulated over decades, the issue still remains unresolved.

In this article, by observing the field-driven metallic to insulating phase transition, we argue that the mechanism of 2D MIT is the percolation in our monolayer MoS₂ and, in general, is not universal. In addition, we observe another phase transition from an insulating to a metallic phase at higher voltage, and we further estimate the activation energy from this transition.

RESULTS AND DISCUSSION

Temperature- and Electric-Field-Dependent Conductivity. For conductivity measurement, we fabricated a monolayer MoS₂ field-effect transistor with electrodes for four-probe measurements, as shown in the inset of Figure 1a.

![Figure 1](image.png)

Figure 1. (a) Electric conductivity ($\sigma$) with respect to the temperature ($T$) at the drain–source voltage $V_{ds} = 0.1$ V under a backgate bias $V_G$ ranging from 10 (bottom) to 80 V (top). The inset shows an optical image of monolayer MoS₂ on the hBN film. (b) $\sigma$ as a function of the electric field ($E \propto V_{ds}$) at $T = 20$ K.

Monolayer MoS₂ was grown by chemical vapor deposition and transferred onto a 15–20 nm thick hexagonal boron nitride (hBN) film to improve the sample quality.$^{17}$ Figure 1a shows the temperature ($T$)-dependent conductivity measured at a drain–source voltage of $V_{ds} = 0.1$ V for a backgate bias $V_G$ ranging from 10 (bottom) to 80 (top) V. Two distinct temperature dependences, $\Delta \sigma/\Delta T > 0$ and $\Delta \sigma/\Delta T < 0$, at a low temperature reveal the occurrence of the MIT around $V_G = 55–60$ V.

Figure 1b shows the electric-field ($E$)-dependent conductivity at $T = 20$ K for the same range of $V_G$ as shown in Figure 1a. Here, $E = V_{ds}/L$ and $V_{ds} = V_{ds} - I_D R_s$, where $L$ is the channel length, $I_D$ is the drain–source current, and $R_s$ is the contact resistance. In this figure, the distinct MIT phenomena are featured in two different field dependences, $\Delta \sigma/\Delta E > 0$ and $\Delta \sigma/\Delta E < 0$ across $V_G = 55–60$ V in the low-field regime. However, the conductivities exhibit minima at $E \approx 0.5$ kV-cm⁻¹ in the metallic regime of $V_G \geq 50$ V and increase again. In general, the increased conductivity with $E$ is inherent for an insulator, caused by the electric-field-enhanced hopping or heating as seen for $V_G < 50$ V. Thus, this nonmonotonic behavior indicates the possible phase transition from metallic to insulating upon increasing $E$ for $V_G \geq 50$ V. In fact, the temperature dependence of the conductivity ensures that the phase of this region (i.e., $E > 0.5$ kV-cm⁻¹ for $V_G \geq 50$ V) is insulating, as we will see later. We ascribe this anomalous behavior to the voltage (or electric-field)-induced localization, which occurs in the regime of the percolation transport.

Anomalous Conductivity and Metal–Insulator Transition. The conductivity crossover for MIT is displayed in temperature dependence as a function of $V_G$ at a given $V_{ds}$ (Figure 2a). The critical backgate bias $V_G$, i.e., $V_G$ at the crossover (arrows), shows the nonmonotonic $V_{ds}$ dependence. As shown in the inset at the bottom of Figure 2a, it initially increases with $V_{ds}$ but starts to decrease after $V_{ds} \approx 0.7$ V. This anomalous behavior implies that the state at a given $V_G$ can be driven to an insulating or a metallic phase by the $V_{ds}$. For example, if we sweep $V_{ds}$ at fixed $V_G = 80$ V along the arrow in the inset, the phase transition occurs twice from metallic to insulating at low voltage and to metallic again at high voltage (Figure 2d for the visualization). This situation is clearly illustrated in Figure 2b. Here, we used $V_{ds}$ instead of $E$ ($= V_{ds}/L$), but it retains the basic features. Considering the temperature dependence, the conductivity exhibits the insulating behavior ($\Delta \sigma/\Delta T < 0$) for the entire experimental field (or voltage) range at $V_G = 30$ V. However, at $V_G \geq 60$ V, three different regimes alternate as $E$ increases, that is, a metallic–insulating–metallic temperature dependence in the conductivity.

We visualize the change of a conductivity in a color-coded plot of a log–log scale for the entire ranges of $V_G$ and temperature (Figure 2c,d at $V_G = 30$ and 80 V, respectively). Figure 2c confirms that the temperature dependence of conductivity at $V_G = 30$ V is insulating for entire values of $V_{ds}$. In contrast, at $V_G = 80$ V, the metallic region apparently appears in low and high voltages (Figure 2d). We suggest the percolation as the origin of this unusual transport property. In particular, in the low-field regime, the decrease of the conductivity as $E$ increases in the insulating phase (e.g., $\sigma$ for $T \geq 40$ K at $V_G = 30$ V in Figure 2b) contrasts the usual insulating behavior and strongly supports this suggestion. This behavior has been experimentally observed in low-compensated semiconductors$^{5,19}$ and interpreted as a result of “dead ends”, that is, the sites distant from neighboring sites, so that the hopping probabilities to them are negligible in the percolation picture$^{20}$ or “returns”, that is, the twisted paths containing the segments where electrons travel along the field.$^9$

In a two-probe measurement, this phenomenon is not seen due to the high contact resistance (Supporting Information Figure S1).
Hopping Transport and Percolation Transition. First of all, to be more specific for the decrease of the conductivity with $E$ in the insulating phase, we illustrate the hopping paths of the electrons trapped by impurities in Figure 3a. In the low field, the electrons that enter the dead ends escape from them easily. However, as $E$ increases, the escape along the field direction becomes more difficult, as shown in Figure 3a, that is, localized in some sense, which results in the decrease of the conductivity. In this regime and at a low temperature, satisfying the condition $qEL_0 \gg k_BT$, where $q$ is the elementary charge, $L_0$ is the correlation length defining the typical size of the closed hopping path (called a cluster), and $k_B$ is the Boltzmann constant, the conductivity is as follows:

$$\sigma \propto \exp(-qEL_0/k_BT)$$

where $\sigma$ is the conductivity, $E$ is the electric field, and $T$ is the temperature.
the MoS2 undergoes the phase transition to an insulating state.

For $V$ rapidly with the increase of $V$, this feature is more pronounced at lower temperature and $G > 40 \text{ V}$, however, the phase remains metallic. However, as shown in Figure 3c for $G = 35 \text{ V}$ and $L_0 \approx 0.33 \mu m$. In the percolation picture, as the carrier density increases to approach the percolation threshold, several clusters merge together to produce the infinite cluster. Thus, $L_0$ diverges at the percolation threshold also. For example, we survey $V_G$ dependence of $L_0$ at $T = 30 \text{ K}$. As shown in Figure 3c for the extracted $L_0$, the increasing tendency of $L_0$ with $V_G$ seems clear for $V_G \leq 40 \text{ V}$. For $V_G > 40 \text{ V}$, however, the fittings (red solid lines) seem disputable because of insufficient data points. In addition, we note that the application of eq 1 becomes inappropriate as $V_G$ is closer to the transition as the hopping model is invalid when the localization length becomes larger than the hopping length. This phenomenon of conductivity drop with $E$ in the insulating phase was predicted to disappear if the temperature is too low because most electrons are localized, so that the density of electrons for hopping is exponentially small. Our observations in Figure 3e are consistent with this prediction, in which the behavior of the conductivity becomes weaker and disappears as the temperature (top figure) or $V_G$ (bottom figure) decreases. 

In monolayer MoS$_2$, we observed this behavior in the sample with a relatively high four-probe field-effect mobility of $\mu_{FE} = (1/C_{ow})(d\sigma/dV_G) \approx 600 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ calculated at $V_{ds} = 0.1 \text{ V}$ and $T = 20 \text{ K}$, where $C_{ow}$ is the geometrical oxide capacitance of 300 nm thick SiO$_2$ (Supporting Information S2) because it requires the large site spacing to have numerous dead ends or returns as this phenomenon has usually been observed in the low compensated semiconductor systems. $\mu_{FE}$ decreases rapidly with the increase of $V_{ds}$ as shown in Figure 3f as it is calculated with the conductivity in the regime of the metallic phase.

In contrast, as it should be in the metallic phase at $V_G \geq 60 \text{ V}$, this feature is more pronounced at lower temperature (Figure 2d). In general, the conductivity decreases as the field increases in the metallic phase because of the Joule heating or hot electron effects, but the phase remains metallic. However, in the present monolayer MoS$_2$, the main driving mechanism for the conductivity decrease in the low-field regime of a metallic phase apparently differs from heating effects because the MoS$_2$ undergoes the phase transition to an insulating state. The same behavior in the insulating phase, that is, the decrease of the conductivity with the field (Figure 3d,e), supports a nonheating effect for such a behavior. This transition is unusual because the electric field usually lowers the potential barrier or promotes the mobility edge, favoring the metallic state. To have this type of phase transition, these delocalizing effects should be overcome by the presence of dead ends or returns in the percolation description. We believe that this is the case for our monolayer MoS$_2$. In other words, as electrons become localized by the field due to the presence of “dead ends” or “returns”, the Fermi level is lower than the percolation threshold at some point to be insulating. As discussed later, an additional phase transition of insulating-to-metallic at a high field is the result of the opposite case, where the delocalizing effects are dominant over the localizing effects. This field-induced insulating-to-metallic phase transition at high field has been reported in other systems.

As the field increases further, the transport becomes more directional (one-dimensional); that is, the electrons gain enough energy to hop to the next distant site. Thus, the conductivity starts increasing with the field strength. In intermediate fields after the minima, the conductivity increases exponentially and is well described by the Poole–Frenkel (PF) effect, where the trap potential barrier varies as follows:

$$\varphi = \varphi_0 - (q^3/\pi\varepsilon_0)^{3/2} E^{1/2}$$

where $\varphi_0$ is the barrier height in the absence of $E$, $\varepsilon$ is the relative dielectric constant, and $\varepsilon_0$ is the dielectric constant of a vacuum. In this case, the conductivity has the form of $\ln \sigma \propto E^{1/2}$. In Figure 3b, the blue curve is the fit to the data with this form, and Figure 3f illustrates the situation of a potential deformation with $E$. The above PF theory fundamentally assumes the long-range Coulombic impurity potentials. Thus, the application of this theory implies the dominant long-range disorder in this sample, and this fact supports the percolation transport. Under higher fields, however, it deviates from the classical PF-type, suggesting that another conduction mechanism is involved. In the presence of significant carrier–carrier interactions, the conduction under high fields is usually described by the Efros and Shklovskii theory, where $\ln \sigma \propto (-E)^{-1/2}$ (ES hopping). The red curve in Figure 3b shows the fit obtained using this model. It appears reasonable to apply this mechanism to monolayer MoS$_2$ because the interaction effects are strong. We note, however, that the fittings by a variable range hopping (VRH) model without the consideration of carrier–carrier interactions, for example, $\ln \sigma \propto$...
\[ (-E)^{-1/3} \]
give similar results. Here, in fact, we cannot determine which is correct. Nevertheless, we point out that ES or VRH appears likely as a result of the narrower and shallower effective barriers between the electron puddles at high fields, so that tunneling through these saddle points starts playing an important role.

These two model fittings (PF and ES here) well describe the field-dependent conductivities for other \( V_G \) values, as well. We define the crossover field \( E_{Qc} \) at which the hopping mechanism switches from PF to ES. As shown in Figure 4a, the values are determined where it deviates from the linear behavior (PF mechanism) in a logarithmic scale with respect to \( E^{1/2} \) at \( T = 20 \) K. For higher fields, \( E > E_{Qc} \), Figure 4b,c demonstrates that it is governed by the ES mechanism; that is, the conductivity is linear in \( \ln \sigma \) versus \( E^{-1/2} \).

**Metal–Insulator Transition at High Field and Activation Energy.** However, the deviations from ES hopping occur at higher fields, \( E > E_{c} \) for \( V_G \gtrsim 50 \) V, as shown in Figure 4b. The temperature dependence of the conductivity in these regions is metallic (Figure 2b). Therefore, we identified \( E_{c} \) as a critical field at which \( \varphi \approx 0 \). From eq 2, the barrier height is then determined as \( \varphi_{c} \approx (q^{2}/\pi\varepsilon\varepsilon_{0})E_{c}^{5/2} \), where \( \varepsilon \approx 4 \) for monolayer MoS\(_{2}\).\(^{28} \) and the result for a different \( V_G \) is shown in the inset of Figure 5. The values are in the range of 30–45 meV for the given range of \( V_G \) and are consistent with the activation energy determined previously from the temperature dependence of conductivity (i.e., the Arrhenius law).\(^{29} \) Here, we neglect the interference effect that possibly exists above the classical percolation threshold, \( \varphi \approx 0 \). If this effect plays a role and a classical threshold is at \( \sim E_{Qc} \), \( \varphi_{c} \) is lower than the estimated values. In this case, \( E_{c} \) may be interpreted as a value corresponding to the mobility edge driven by the electric field, which was theoretically predicted.\(^{30–32} \)

**Phase Diagram.** We have discussed phase transitions driven by the electric field in monolayer MoS\(_{2}\). Figure 5 shows the summarized phase diagram in \( V_G \) versus \( E \). Here, \( E_{M} \) denotes the field at which the conductivity is a minimum. The real critical field for the phase transition from metallic to insulating is lower than \( E_{M} \). There is an argument that the electric field is ineffective to weak localization, as it does not break the time-reversal symmetry. In contrast, as previously mentioned, numerous theories predict the emergence or enhancement of the mobility edge by the electric field.\(^{30–32} \) In either case, the phase transition from metallic to insulating with a field in monolayer MoS\(_{2}\) suggests that the insulating phase is not an Anderson insulator. Mott insulators are also known to experience electric-field-driven breakdown.\(^{24,33,34} \)

This certifies our interpretation of the percolation rather than the QPT being the origin for the MIT phenomena in this monolayer MoS\(_{2}\).

**Power Law Behavior of Conductivity in Percolation Transition.** The percolation transport in monolayer MoS\(_{2}\) has been examined with several different measurements such as capacitance,\(^{28} \) noise,\(^{35} \) current measurements\(^{36} \) (see Supporting Information for similar current–voltage scaling analyses), etc. Another mostly applied inspection for percolation has been made through the carrier-density-dependent conductivity at zero temperature. The percolation theory predicts the relationship \( \sigma(T = 0) \sim (n - n_{c})^{\mu} \), with the exponent value \( \mu = 4/3 \) for a 2DS,\(^{21} \) where \( n \) and \( n_{c} \) are the carrier density and the critical carrier density, respectively. Assuming that \( n = C_{o}(V_{G} - V_{TH})/q \) holds, we have \( (n - n_{c})^{\mu} \sim (V_{G} - V_{c})^{\mu} \), where \( V_{TH} \) is the threshold voltage and \( V_{c} \) is the critical backgate bias.

For the fitting, we used \( V_{c} \) for the fitting parameter instead of \( V_{G} \), Figure 6a shows the conductivity and fittings using the above equation for chosen temperatures. To minimize the thermal excitation effect, we take the highest \( V_{c} \) region into account for fittings, giving unsurprisingly poor fittings to the region near \( V_{c} \approx 55 \) V. The obtained fitting parameters \( \mu \) and \( V_{c} \) are shown in Figure 6b. At the lowest temperature \( T = 20 \) K, \( \mu \approx 1.32 \), and \( V_{c} \) approach \( V_{c} \) consistent with the percolation.

The 2D MIT phenomenon has been observed in many other systems, typically Si-MOSFETs and GaAs/AlGaAs. The dominant disorder in these two systems is known to differ: short-range for the former and long-range for the latter. It has been speculated that this difference is the underlying origin for the different reports on the mechanism,\(^{37} \) QPT in Si-MOSFETs,\(^{11,13} \) and percolation in GaAs/AlGaAs.\(^{15,38,39} \) On the other hand, opposite conclusions based on the results from different aspects have also been reported.\(^{40,41} \) Presently, it appears uncertain that any exclusive experimental result can fully elucidate the 2D MIT phenomena. The scaling behavior of the conductivity for temperature is, in general, used to demonstrate QPT, but it is not so experimentally trivial to implement all typical processes indisputably because of the finite temperature effects ignoring possible weak localization effects,\(^{16} \) the difficulty in determination of the critical density, heating effects, the possible trap effects,\(^{12} \) etc. (Supporting Information S4 for scaling analysis of the present monolayer MoS\(_{2}\).) On the other hand, the experimental value of \( \mu \) close to 4/3 alone does not guarantee the percolation transition.

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**Figure 5.** Phase diagram. M and I represent the metallic and insulating phases, respectively. The inset shows the potential barrier height estimated according to the critical field \( E_{c} \).

**Figure 6.** Power law fits to the conductivity. (a) \( \sigma \) vs \( V_{c} \) for chosen temperatures. Solid lines are power law fits to the conductivity with \( (V - V_{c})^{\mu} \). (b) Derived exponent \( \mu \) (top) and fitting parameter \( V_{c} \) as a function of temperature (bottom).
because the theoretical value of $\mu$ is not known for QPT in 2D. However, if $\mu$ is far from 4/3, it may be reasonable to say that it is not consistent with a percolation. In this study, $\mu$ was extracted at rather high temperatures. However, we expect that true $\mu$ is not significantly out of the tolerable range from 4/3 as our lowest temperature of 20 K is much lower than the Fermi temperature of $\sim$150 K and near the Dingle temperature of $T_D = qh/(2mk_BT)$ $\lesssim$ 22 K, where $m$ is the effective mass and $h$ is the reduced Planck constant.

CONCLUSION

The conductivity in monolayer MoS$_2$ exhibits diverse features under an electric field. In particular, the field-induced metal-to-insulator transition and the decrease of the conductivity with a field in the insulating phase in the low-field regime strongly suggest the percolation as the origin of the carrier-density-tuned MIT in monolayer MoS$_2$. This conclusion opposes the interpretation of the quantum phase transition based on the reduced Plank constant.

METHODS

Device Fabrication and Electrical Transport Measurement

Monolayer MoS$_2$ was grown by the chemical vapor deposition method and transferred onto a multilayer (15 nm thick) hBN substrate. To pattern the electrodes, PMMA A4 was spin-coated (3000 rpm, 30 s), and then the electron beam lithography was performed, followed by Cr/Au (2/60 nm) evaporation in high vacuum ($\sim$10$^{-6}$ Torr). The dimensions of the monolayer MoS$_2$ device are 7.7 $\mu$m in length and 6.2 $\mu$m in width. Four-probe electrical measurements were performed in high vacuum ($\sim$10$^{-6}$ Torr) using a cryostat (PPMS, Quantum Design, Inc.) with a characterization system (B1500A, Keysight Technologies).

ASSOCIATED CONTENT

1. Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nano.9b00755.

$I_{th}$ vs $V_G$ and $I_{th}$ vs $V_{ch}$ in two-probe measurements, temperature-dependent conductivity in monolayer MoS$_2$, $I_{th}$ scaling for $V_{ch}$, and temperature scaling (PDF)

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Notes

The authors declare no competing financial interest.

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