Metal-insulator transition in a transition metal dichalcogenide: Dependence on metal contacts

Y Shimazu¹, K Arai and T Iwabuchi

Department of Physics, Yokohama National University
79-5 Tokiwadai, Hodogaya, Yokohama 240-8501, Japan

¹E-mail: yshimazu@ynu.ac.jp

Abstract. Transition metal dichalcogenides are promising layered materials for realizing novel nanoelectronic and nano-optoelectronic devices. Molybdenum disulfide (MoS₂), a typical transition metal dichalcogenide, has been extensively investigated due to the presence of a sizable band gap, which enables the use of MoS₂ as a channel material in field-effect transistors (FET). The gate-voltage-tunable metal–insulator transition and superconductivity using MoS₂ have been demonstrated in previous studies. These interesting phenomena can be considered as quantum phase transitions in two-dimensional systems. In this study, we observed that the transport properties of thin MoS₂ flakes in FET geometry significantly depend on metal contacts. On comparing Ti/Au with Al contacts, it was found that the threshold voltages for FET switching and metal–insulator transition were considerably lower for the device with Al contacts. This result indicated the significant influence of the Al contacts on the properties of MoS₂ devices.

1. Introduction

Molybdenum disulfide (MoS₂) is a promising semiconducting material owing to its ultrathin structure that enables scaling of the MoS₂ devices to less than several nanometers [1-4]. From the perspective of fundamental physics, the superconductivity [5,6] and metal–insulator transition (MIT) [7-9] observed in MoS₂ have been given much attention, because these may be considered as quantum phase transitions in two-dimensional systems. In previous studies, Au [7,9] and Ti/Au [6,8] (Ti is the bottom layer) have been widely used as the contact metals for electrodes. The dependence of the transport properties on the contact metal has rarely been investigated. Particularly, the influence of the metal contacts on MIT has not been reported. We studied the transport properties of multilayer MoS₂ flakes with Au and Al contacts in a field-effect transistor (FET) geometry from room temperature (290 K) to 13 K. The reports on the transport properties of transition metal dichalcogenides that are contacted by Al have been very limited [10-12]. We found that the temperature dependences of the sheet resistance, relevant to the MIT, are different for the devices with the two types of contacts. The MIT at low temperatures was observed in back-gated FETs with Al contacts.

¹ To whom any correspondence should be addressed.
2. Experimental
MoS₂ flakes were exfoliated repeatedly from a bulk crystal (SPI Supplies) using adhesive tape [13]. Subsequently, thin flakes were transferred to a Si substrate with a 270-nm-thick SiO₂ layer via a gel sheet [14]. The highly n-doped Si substrate was used as a back gate. The source and drain electrodes were fabricated using photolithography and electron-beam deposition of metals. In the devices with Au contacts (referred as Au-contacted devices), Ti(12 nm)/Au(75 nm) were used as contact metals, where Ti was the bottom layer. In the devices with Al contacts (referred as Al-contacted devices), Al (90 nm) was used as a contact metal. For the Al-contacted devices, we deposited Ti (15 nm)/Au (50 nm) layers on the Al pads for connection of Au wires to the contact pads, where the Ti/Au layers were away from the MoS₂ flakes by more than 350 μm. The photographs of the Al-contacted device are shown in figure 1. We measured the drain–source current \( I_d \) as a function of drain–source voltage \( V_d \) and back-gate voltage \( V_g \).

3. Results and discussion
The \( I_d–V_d \) curves at room temperature for the Au-contacted and Al-contacted devices are shown in figures 2(a) and (b), respectively. The thicknesses of the MoS₂ flakes in these devices are 16 nm, as determined by an atomic force microscope. Both the devices exhibit n-channel FET characteristics; at low values of \( V_g \), the off-state is realized. As shown in the figures, the threshold value of \( V_g \) for the FET switching is lower in the Al-contacted device than that in the Au-contacted device. In the on-state, the \( I_d–V_d \) curve is nearly linear for the Au-contacted device, while that for the Al-contacted device is nonlinear. The nonlinearity reflects the significance of Schottky barrier between the Al contacts and MoS₂ flakes. This result implies that the Schottky barrier associated with Al contacts is higher and/or thicker than that associated with Au contacts. This observation is inconsistent with that indicated by the work functions of Al (4.54 eV) and Au (5.40 eV) [15]; from the Schottky-Mott rule for the Schottky barrier height, the barrier height for Al should be lower than that for Au [10]. Existence of
high-resistance Schottky barrier for Al contact has been reported in the literature [11] and is attributed to the lack of d-orbitals in Al, which leads to a small overlap of the electron orbitals with the d-orbitals in MoS2 [11].

In contrast to the $I_d$–$V_g$ curve, the four-terminal current–voltage curves were nearly linear for both types of the devices from room temperature to 13 K. The sheet resistance is derived from the four-terminal measurements. The sheet resistances $R_s$ (at various values of $V_g$) as functions of temperature for the Au-contacted and Al-contacted devices are shown in figures 3(a) and (b), respectively. The temperature dependence of $R_s$ indicates a qualitative difference between the two types of devices. For the Au-contacted device, the temperature coefficient of $R_s$ at room temperature, $dR_s/dT$, is negative for $V_g < 27$ V and positive for $V_g > 27$ V. However, the insulating behavior ($dR_s/dT < 0$) is observed at lower temperatures for all the values of $V_g$. In contrast, for the Al-contacted device, the temperature dependence at lower temperatures (down to 13 K) shows a MIT with critical point at $V_g \sim -10$ V, while that near room temperature shows a metallic behavior ($dR_s/dT > 0$).

The difference, as shown in figures 3(a) and (b), can be explained by assuming that the carrier density in the Al-contacted device is higher than that in the Au-contacted device. This assumption is consistent with the difference in the threshold value of $V_g$ for FET switching in the devices with different contacts. The higher carrier density in the Al-contacted device may possibly be caused by electron transfer from Al to MoS2, which is expected on the basis of the work functions of Al (4.54 eV) and Au (5.40 eV). For the Al-contacted devices, the carrier density in MoS2 at equilibrium is influenced by the work function of Al, and not by the significant Schottky barrier at the Al contacts. We found that the presence of Ti/Au layer on the Al contact pads has a significant influence on the transport properties of the devices. Further study is required to elucidate the effects of Al contacts on the devices.

MITs in MoS2 have been reported in the literature. In [7], MIT in monolayer MoS2 is reported in a top-gated device using HfO2 as a top-gate dielectric. Only insulating behavior is observed for back-gate voltages up to 40 V in this device. In [8,9], MITs in back-gated monolayer MoS2 are reported, wherein the thicknesses of the SiO2 dielectric are similar to that in our devices. For multilayer MoS2 flakes, MIT and field-induced superconductivity are reported in [5,6], where significant electrostatic
carrier doping is achieved using ionic liquids. In [5-9], the critical resistance of MIT is observed to be close to the quantum resistance $h/e^2$ (~25.8 kΩ) with an exception of [5] in which the critical sheet resistance is observed to be between 100 Ω and 1 kΩ. The quantum resistance is the most relevant unit of resistance in the analysis of MIT in two-dimensional electron systems [16]. In our results (shown in figures 3(a) and (b)), the critical resistance of MIT in the Al-contacted device is ~10 kΩ, and that of the MIT-like behavior near room temperature in the Au-contacted device is ~60 kΩ. Therefore, the critical resistance (~10 kΩ) of MIT observed in our study is near the quantum resistance, which is consistent with other reports. The critical resistance being close to the quantum resistance is expected from the Ioffe–Regel criterion for MIT, $k_F l_e \sim 1$, where $k_F$ and $l_e$ are the Fermi wave vector and mean free path of electrons, respectively. It has been revealed that this criterion is consistent with experimental data of MIT [7].

**Figure 3.** Sheet resistance of MoS$_2$ FETs with Ti/Au contacts (a) and Al contacts (b) as a function of temperature for various values of $V_g$; the resistance decreases with increasing $V_g$ at all the temperatures. MIT is observed at low temperatures for the Al-contacted device.

4. Conclusion
We examined the transport properties of multilayer MoS$_2$ flakes with Au and Al contacts in a back-gated FET geometry from room temperature to 13 K. For the Al-contacted devices, MIT was observed at lower temperatures (down to 13 K), with a critical resistance close to the quantum resistance, while the Au-contacted devices were insulating in the same range of temperature and for $V_g$ up to 40 V. This result implies that the MIT depends on the metal contacts and that the Al contacts induce higher carrier density in MoS$_2$ flakes.
Acknowledgments
This work was supported by JSPS KAKENHI Grant Number 15K13497.

References
[1] Wang Q H, Kalantar-Zadeh K, Kis A, Coleman J N and Strano M S 2012 Nat. Nanotechnol. 7 699–712
[2] Ganatra R and Zhang Q 2014 ACS Nano 8 4074–4099
[3] Tong X, Ashalley E, Lin F, Li H and Wang Z M 2015 Nano-Micro Letters 7 203–218
[4] Wang H, Yuan H, Hong S S, Li Y and Cui Y 2015 Chem. Soc. Rev. 44 2664–2680
[5] Taniguchi K, Matsumoto A, Shimotani H and Takagi H 2012 Appl. Phys. Lett. 101 042603
[6] Ye J T, Zhang Y J, Akashi R, Bahramy M S, Arita R and Iwasa Y 2012 Science 338 1193–1196
[7] Radisavljevic B and Kis A 2013 Nat. Mater. 12 815–820
[8] Baugher B W H, Churchill H O H, Yang Y and Jarillo-Herrero P 2013 Nano Lett. 13 4212–4216
[9] Schmidt H, Wang S, Chu L, M Toh, Kumar R, Zhao W, Castro Neto A H, Martin J, Adam S, Ozyilmaz B and Eda G 2014 Nano Lett. 14 1909–1913
[10] Walia, S. et al. 2013 Appl. Phys. Lett. 103 232105
[11] Liu W, Kang J, Sarkar D, Khatami Y, Jena D and Banerjee K 2013 Nano Lett. 13 1983–1990
[12] Kwon J, Lee J Y, Yu Y J, Lee C H, Cui X, Hone J and Lee G H 2017 Nanoscale 9 6151–6157
[13] Novoselov K S, Geim A K, Morozov S V, Jiang D, Zhang Y, Dubonos S V, Grigorieva I V and Firsov A A 2004 Science 306 666–669
[14] Castellanos-Gomez A, Buscema M, Molenaar R, Singh V, Janssen L, van der Zant H S J and Steele G A 2014 2D Materials 1 011002
[15] Skriver H L and Rosengaard N M 1992 Phys. Rev. B 46 7157–7168
[16] Kravchenko S V and Sarachik M P 2004 Rep. Prog. Phys. 67 1–44