Electron accumulation-type Ohmic contact for MoS$_2$ field-effect transistor

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Abstract

The formation of an Ohmic contact at a metal/two-dimensional (2D) semiconductor interface is a critical step for the future development of high-performance and energy-efficient electronic and optoelectronic applications based on semiconducting transition-metal dichalcogenides$^1$. The deposition process of metals at high thermal energy introduces crystalline defects in 2D semiconducting layers, leading to an uncontrollable Schottky barrier height regardless of work function of a metal and high contact resistance$^{2,3}$. Here, we report the fabrication of Ohmic contacts by evaporation of indium (In) at a relatively low thermal energy onto molybdenum disulfide (MoS$_2$), resulting in a van der Waals (vdW) In/MoS$_2$ accumulation-type contact with a metal-induced electron doping density as $\sim10^{12}$ cm$^{-2}$. We
show that the transport at the In/MoS\textsubscript{2} accumulation-type contact is dominated by the field-emission mechanism over a wide temperature range from 2.4 to 300 K and at a carrier density as low as \(~10^{12}\) cm\(^{-2}\) for a few-layered MoS\textsubscript{2} device. In this case, the contact resistance reaches 0.6 k\(\Omega\) \(\mu\)m at cryogenic temperatures. These results pave a practically available path for fabricating Ohmic MoS\textsubscript{2} contacts for high-performance electronic and optoelectronic applications.

Layered semiconducting transition-metal dichalcogenides (TMDCs) such as MoS\textsubscript{2}, WSe\textsubscript{2} and MoTe\textsubscript{2} have been intensively studied for use in semiconducting electronic and optoelectronic devices\textsuperscript{4}. Because each atomic layer in such bulk TMDCs is coupled with neighboring layers through van der Waals (vdW) interactions, mono- or few-layer flakes of these materials can be deposited onto a substrate via a mechanical exfoliation method\textsuperscript{5}, which has opened a new platform, i.e., two-dimensional (2D) semiconducting electronics, for the future development of low-power, high-performance electronics\textsuperscript{4,6}.

The inert character of TMDC atomic layers enables the fabrication of vertical vdW heterostructures as new functional electronics and optoelectronics\textsuperscript{7,8,9}. However, such inert character is known to inhibit direct doping by atomic substitution\textsuperscript{1}. In Si-based electronics, a strong doping process beneath the metal contacts by atomic substitution results in a narrow Schottky barrier, which in turn leads to a reliable Ohmic contact with field-emission (or tunneling) current as the dominant transport mechanism at the contacts\textsuperscript{10}. In the case of devices based on MoS\textsubscript{2} as the TMDC material, efforts to identify metals with appropriate work functions (\(\Phi_{\text{metal}}\)) compared with the affinity of MoS\textsubscript{2} (monolayer MoS\textsubscript{2}: \~4 eV, multilayer MoS\textsubscript{2}: \~4.3 eV), such as Sc (\(\Phi_{\text{Sc}} = 3.5\) eV) and Ti (\(\Phi_{\text{Ti}} = 4.3\) eV), to reduce the Schottky barrier height have not been effective.
because of Fermi-level pinning (FLP)\textsuperscript{11,2}. Researchers have explored various approaches to overcoming this problem, including molecular doping\textsuperscript{12}, tunnel-barrier insertion\textsuperscript{13,14}, fabrication of graphene contacts\textsuperscript{15,16}, and phase changes\textsuperscript{17}. The several results of these studies have shown that an Ohmic contact with thermionic emission at room temperatures plays a dominant role in contact transport in most cases. In that case, the contact resistance increases as the temperature is lowered because of the suppression of the thermionic currents. In this view, the contact transport obeying the field-emission mechanism based on an accumulation-type contact is expected to support the Ohmic contact for a wide temperature range including a cryogenic environment\textsuperscript{14}.

Here, we report the realization of the accumulation-type Ohmic contact having a contact resistance, $R_c W$, of $\leq 1$ k$\Omega$ $\mu$m at cryogenic temperatures of $T < 100$ K for direct-evaporated indium ($\text{In}$) contacts having a relatively low thermal energy on few-layered MoS$_2$, resulting in an “vdW In/MoS$_2$ interface” without crystalline defects of MoS$_2$ at the interface. This work reveals that the $R_c W$ behaviors at various temperatures and carrier densities ($n_c$) are dominated by the sheet resistance of MoS$_2$ ($R_{\text{sh}}$), rather than by the specific contact resistivity ($\rho_c$), which is governed by the field-emission process. We estimated the Schottky barrier height for In/few-layer MoS$_2$ contacts as $\leq 10$ meV based on the field-emission mechanism, reflecting FLP-free contacts. As expected in an accumulated contact with $\Phi_{\text{In}} \approx 4.1$ eV, there was a substantial electron transfer from In to MoS$_2$ at the contact region, which was confirmed by Raman spectra.

**Characterization of van-der-Waals In/MoS$_2$ interface**

We fabricated MoS$_2$ field-effect transistors (FETs) on hexagonal boron nitride (hBN) flakes, where the hBN flakes were deposited onto a 300-nm-thick SiO$_2$/Si substrate by mechanically exfoliation. We then transferred a few-layer MoS$_2$ flake (HQ-graphene, Inc.) onto a selected hBN
flake\textsuperscript{15,18}. For the electrical measurements, we deposited 100 nm-thick In electrodes across the MoS\textsubscript{2} channel (Fig. 1a), where the substrate holder was cooled to \(~100\) K by liquid nitrogen flowing through it\textsuperscript{19}. Figure 1b shows a cross-sectional transmission electron microscopy (TEM) image of the In/few-layered MoS\textsubscript{2} junction of a separately prepared sample, which clearly shows an atomically separated interface between In and MoS\textsubscript{2} layers without any metal invasion into the MoS\textsubscript{2} layers. The left and right panels of Fig. 1c show optical images before and after a mechanical process to remove the deposited In metals from a top of MoS\textsubscript{2} via adhesive tape, respectively. These images show that the original MoS\textsubscript{2} under the In remained intact after the In was removed, indicating the formation of an In/MoS\textsubscript{2} vdW interface\textsuperscript{3}. Whereas FLP can originate from crystal-lattice disorder and defect-induced gap states that occur during the deposition process of a metal at high thermal energy because of its high evaporation temperature\textsuperscript{2,3}, our vdW interface with In deposited at a relatively low thermal energy could provide a FLP-free contact. For instance, whereas the evaporation temperature of Au at \(10^{-8}\) Torr is \(~800\) °C, only \(~480\) °C is required for the evaporation of In at the same pressure. In electrical measurements, to apply an electric field to the MoS\textsubscript{2} channel, the highly \(p\)-doped Si substrate was biased by a back-gate voltage (\(V_G\)). All measurements were performed in a cryostat (PPMS, Quantum Design, Inc.) with a base \(T\) of 2.5 K.

**Basic electrical properties of a few-layer MoS\textsubscript{2} field-effect transistor**

Figure 1a shows a photograph of a MoS\textsubscript{2} FET on a 22 nm-thick hBN flake (6L-MoS\textsubscript{2} device). The number of MoS\textsubscript{2} layers was estimated as \(n = 6\) (see Fig. S1 in Supplementary). The multiple electrodes for the MoS\textsubscript{2} flake with different intervals between two neighboring electrodes were designed to measure the contact resistance via the transfer-length method (TLM)\textsuperscript{17}, i.e., four
FETs with different channel lengths ($L_1 = 0.5 \, \mu m$, $L_2 = 1 \, \mu m$, $L_3 = 1.5 \, \mu m$, and $L_4 = 2 \, \mu m$ from the left channel in the region indicated by a dashed box). Here, the widths ($W$) of all channels were identical at $2 \, \mu m$.

Figure 1d shows the two-probe conductance as a function of the back-gate voltage ($G–V_G$) of the $L_2$-FET with a source–drain voltage ($V_{SD}$) of $30 \, mV$ at various temperatures. The conductance decreased for negatively decreasing $V_G$ and reached zero near $V_G \approx 0 \, V$ throughout the investigated temperature range, which indicates that the carriers were electrons. The two-probe conductance increased with decreasing $T$ at a given $V_G$ for $V_G > 10 \, V$, i.e., the device exhibited metallic behavior. However, the opposite behavior was observed near a depletion region of $0 < V_G < 10 \, V$, representing insulating character. These behaviors are consistent with the current–voltage ($I–V_{SD}$) curves for various $V_G$ values at $T = 2.4 \, K$ in Fig. 1e. For instance, the $I–V_{SD}$ curves for $V_G > 10 \, V$ and $0 < V_G < 10 \, V$ show linear and nonlinear characteristics, respectively. The four-probe measurements for the $L_2$ channel also showed a similar $V_G$ value for the metal–insulator crossover location (see Fig. S2 in Supplementary). This result indicates that the crossover behavior is mainly determined by the transport in the MoS$_2$ channel, not in the contact part.

Figure 1f shows the field-effect mobility ($\mu$) as a function of $T$, as obtained from the two-probe (opened circles) and four-probe (closed squares) measurements. The mobility was obtained at the local maximum location in the $\mu–V_G$ curves (see Fig. S3 in Supplementary). For $T > 100 \, K$, in both cases, the data were fitted with the relation $\mu(T) = \mu_0 T^{-\alpha}$ (where $\alpha = 2.2$), as shown by the dashed line. This $\alpha$ value is similar to the expected value for bulk MoS$_2$ ($\alpha = 2.5$) with optical phonon scattering as the dominant scattering mechanism$^{20,15}$. At room $T$, $\mu \approx 50 \, cm^2V^{-1}s^{-1}$ for both cases. However, when $T < 20 \, K$, $\mu$ became saturated at 1200 and 3200 $cm^2$
V$^{-1}$ s$^{-1}$ with decreasing temperature for the two- and four-probe measurements, respectively. Saturation behavior in the low-$T$ region has been known to occur when the impurity scattering plays a dominant role and the phonon effect is suppressed$^7$.

**Contact resistance at In/MoS$_2$ contacts**

On the basis of the TLM (see Fig. S4 in Supplementary) with multiple channels (Fig. 1a), we extracted the contact resistance ($R_c W$) as a function of $V_{G-th}$ ($= V_{G} - V_{th}$) of a 6L-MoS$_2$ device at representative temperatures; the results are shown in Fig. 2a. We note that the obtained contact resistance includes a serial resistance of In electrodes. At a given temperature, $R_c W$ decreased with increasing $V_{G-th}$. The contact resistance is given by$^1$

$$R_c W (n_e, T) = \sqrt{R_{sh}(n_e, T) \rho_c(n_e, T)}$$

(1)

which is only valid for $L_c \gg L_T$. Here, $L_c$ (= 1 μm) and $L_T$ (= $\sqrt{\rho_c / R_{sh}}$) are the contact length and transfer length representing the average distance that charge carriers flow in a semiconductor beneath the contact before they totally transport to the electrode, respectively. Figure S4 shows that our device satisfied this condition with $L_T \approx 0.1$ μm. Equation (1) implies that $R_c W$ decreases with increasing $n_e$ because both $R_{sh}$ and $\rho_c$ generally decrease with increasing $n_e$. However, $R_c W$ increased with increasing $T$ at a fixed $V_{G-th}$ in the $V_G$ range investigated in Fig. 2a, whereas the thermionic emission mechanism with the Schottky barrier at the contact predicts that $R_c W$ increases with decreasing $T$ because of suppression of the thermionic emission$^{21}$.

Figure 2b shows $R_c W$ as a function of $T$ at $V_{G-th} = 45$ V. The contact resistance decreased from 2.3 to 0.6 kΩ μm when the $T$ was decreased from room $T$ to 2.4 K. This behavior has been
reported for graphene/MoS$_2$\textsuperscript{15} and Pd/graphene contacts\textsuperscript{22} in several previous works; it is explained as evidence of the absence of thermionic emission for the contact transport mechanism. Figure 2c shows $R_{sh}$ vs $V_{G-th}$ for the $L_2$ channel at various temperatures, which was also obtained by the TLM (see Fig. S4 in Supplementary). The behavior of $R_{sh}$, which increased with decreasing $V_{G-th}$ at all of the investigated temperatures, is mainly due to decreasing $n_e$. Figure 2d shows $R_{sh}$ as a function of $T$ at $V_{G-th} = 45$ V, where $R_{sh}$ decreased with decreasing $T$. This metallic behavior was observed until $V_{G-th} \approx 15$ V.

**Transport mechanism at In/MoS$_2$ contacts**

We next elucidated which component among $R_{sh}$ and $\rho_c$ predominantly determines the contact resistance at the In/MoS$_2$ vdW contact. The scattered squares in Fig. 3a show $R_cW$ as a function of $n_e$, which was estimated from the relation $n_e = \left( e\mu R_{sh}^{4P} \right)^{-1}$, where $R_{sh}^{4P}$ and $\mu$ were obtained from four-probe measurements. We included another reported result (solid curves) obtained with graphene(Gr)/4L-MoS$_2$ ($n = 4$) contact\textsuperscript{15} for comparison. In this case, the graphene functions as a work-function-controllable contact material, which leads to a lower contact resistance, i.e., $R_cW \approx 1$ kΩ µm at $n_e > 4 \times 10^{12}$ cm$^{-2}$ and $T \approx 10$ K (see the dashed oval in Fig. 3a). Although both the Gr-and In-contact MoS$_2$ devices gave a similar minimum $R_cW$ at cryogenic temperatures, the transport mechanisms at the contacts differ from each other. Near room $T$, the slopes of the $R_cW$–$n_e$ plot do not change for $10^{12} < n_e < 10^{13}$ cm$^{-2}$ in either case. In the case of the In/MoS$_2$ contact, $R_cW$ decreased with decreasing $T$ for the examined $n_e$ range, representing the field emission (or tunneling) for the Schottky barrier for all examined $T$ and $n_e$ ranges. However, the $R_cW$–$n_e$ curves obtained at $T = 12$ and 250 K for the Gr/4L-MoS$_2$ device suggest that the left and right sides with
respect to \( n_e \approx 2 \times 10^{12} \text{ cm}^{-2} \) followed the thermionic and field emissions at the contact, respectively. At \( T = 12 \text{ K} \), although the \( R_c W \) of \(~1 \text{ k}\Omega \mu\text{m} \) was relatively insensitive to the variation of \( n_e \) in the range from \( 4 \times 10^{12} \) to \( 7 \times 10^{12} \text{ cm}^{-2} \) in the dashed oval, it rapidly changed from \( 1 \text{ k}\Omega \mu\text{m} \) to \( 6 \text{ k}\Omega \mu\text{m} \) when \( n_e \) decreased from \( 3 \times 10^{12} \text{ cm}^{-2} \) to \( 1.5 \times 10^{12} \text{ cm}^{-2} \), as shown in the solid oval.

For the In/6L-MoS\(_2\) device, such behavior was not observed.

To clarify the difference between the two cases, we plotted \( R_c W \) as a function of \( R_{sh} \) in Fig. 3b. The slope of the data in the solid oval of Fig. 3b, for \( T = 12 \text{ K} \) (solid green curve), which corresponds to the solid oval in Fig. 3a, followed a relation of \( R_c W \propto R_{sh}^{y} \) with \( y = 7 \) (solid red line), which deviates substantially from the expected value of \( y = 0.5 \) based on equation (1) with an assumption of constant \( \rho_c \) (see the two dashed black lines in Fig. 3b). This result implies that \( \rho_c \), depending on \( n_e \), plays a dominant role in determining \( R_c W \) in this region. For instance, with increasing \( n_e \) or \( V_G \) from the \( V_{th} \), the width of the energy barrier at the contacts to be overcome for electron flow rapidly narrows, resulting in lowering of the \( \rho_c \).\(^{11} \) As a result, \( y \) becomes much greater than 0.5. With increasing \( n_e \) for the Gr/MoS\(_2\) contact, \( R_c W \) becomes saturated for \( n_e > 4 \times 10^{12} \text{ cm}^{-2} \), as shown in the dashed oval in Fig. 3a, where the field emission becomes a dominant factor affecting the contact transport. In this case, the slope of the red dashed line in the corresponding dashed oval in Fig. 3b is less than 0.5, i.e., \( y = 0.2 \). Our In/6L-MoS\(_2\) device with In contact also showed a \( y \) less than 0.5 for \( 2.4 \leq T \leq 100 \text{ K} \) in Fig. 3b. In this temperature region, we thus consider that the field emission plays a dominant role in the vdW contact transport in the In/6L-MoS\(_2\) device.

In our In/6L-MoS\(_2\) device (Fig. 3b), \( R_{sh} \) varies in the range from 1 to 80 \( \text{k}\Omega \) when \( \rho_c \) only varies from \( 0.5 \times 10^{-6} \) to \( 5 \times 10^{-6} \Omega \text{ cm}^2 \), as shown by two dashed lines for \( R_c W \) changing from 0.6
to ~3 kΩ μm. This result indicates that $R_{sh}$ plays a dominant role in determining the $R_LW$ in the field-emission region. A value less than $\gamma = 0.5$ in the field-emission region of the low-$T$ region could be due to the change in $R_{sh}$ under the In (or Gr) contact compared with the original $R_{sh}$ of the MoS$_2$ channel$^1$. Finally, $\gamma$ becomes 0.5 near room $T$ for both devices where $\rho_c$ becomes nearly constant at $\sim 2 \times 10^{-6} \, \Omega \, \text{cm}^2$ with changing $R_{sh}$ and $n_e$. This result indicates that $R_{sh}$ under the contact approaches that of the MoS$_2$ channel with increasing $T$.

**Accumulation-type contacts at In/MoS$_2$ interface**

To evaluate the Schottky barrier height ($\phi_{SB}$) at a metal/semiconductor, it requires measuring the activation energy at the contacts in the thermionic emission region$^{1,11}$, where the voltage drop should mainly occur at the contacts. The inset of Fig. 4b shows an atomic force microscopy (AFM) image of a 1L-MoS$_2$ device with four electrodes prepared on a 40 nm-thick hBN flake (also see Fig. S5 in the Supplementary). Figure 4a shows $G$–$V_G$ curves for various temperatures, where the $L = 0.9 \, \mu$m channel, as indicated by the dashed box in the inset of Fig. 4b, was measured via the two-probe measurement including the contact resistance. In this case, the metal–insulator crossover voltage was found at a relatively high $V_G$ ($\sim 65 \, \text{V}$) compared with those of the 6L-MoS$_2$ devices$^{23}$. $G$ increased with increasing $T$ for $T < 130 \, \text{K}$ at $V_G < 65 \, \text{V}$ and decreased for $T > 200 \, \text{K}$ over the examined $V_G$ range. It is important to know where the insulating behavior mainly originates from, i.e., contact or channel. Figure 4b shows the four-probe $G$–$V_G$ curves for the same channel excluding the contact resistance at the same temperatures in Fig. 4a, where the metal–insulator crossover voltage was also located at $V_G \sim 65 \, \text{V}$ with nearly the same conductance range to the two-probe scheme in Fig. 4a at a corresponding temperature. This indicates that
the insulating behavior at \( V_G < 65 \text{ V} \) is mainly contributed by the MoS\(_2\) channel, not the contact parts, thus, it is not feasible to estimate \( \phi_{SB} \) by using data in Fig. 4a.

We note that both Fig. 1d and Fig. 4a show that the threshold \( V_G \) for few and monolayer MoS\(_2\) is located at \( V_G \geq 0 \text{ V} \), which indicates that the Fermi energy of MoS\(_2\) is near the conduction-band edge in the energy gap. Since the electron affinity of MoS\(_2\) (\( \chi_{\text{MoS}_2} \)) ranges from 4 eV (mono-layer MoS\(_2\)) to 4.3 eV (multilayer MoS\(_2\)), the possible band alignment of at the interface is suggested by two accumulation-type contacts of \( \Phi_{\text{In}} < \chi_{\text{MoS}_2} \) and \( \Phi_{\text{In}} > \chi_{\text{MoS}_2} \) with \( \Phi_{\text{In}} \approx 4.1 \) eV. Figure 5a describes the alignment of bands before (left panel) and after (right panel) contact considering a case of \( \Phi_{\text{In}} \geq \chi_{\text{MoS}_2} \) considering the threshold \( V_G \) location. After contacting, there exists a vdW gap between the In and MoS\(_2\). The region ‘B’ corresponds to the accumulation region of electron carriers at the In-contacted MoS\(_2\) region (see Fig. 5b), where red ‘minus’ signs indicates the accumulated electrons. The band of MoS\(_2\) recovers to the original band (region ‘D’) through the region ‘C’.

To evaluate the electron accumulation at an In-contacted region in MoS\(_2\), we used the Raman spectroscopy to analyze MoS\(_2\) regions covered and not covered by an In electrode. The \( A_{1g} \) phonon peak of MoS\(_2\) is known to exhibit a red shift and its width is known to broaden upon electron doping\(^{24}\). Figure 5c and d show optical images of 1L- and 2L-MoS\(_2\) (indicated by dashed black lines), respectively, prepared on SiO\(_2\), and partially covered with 5 nm-thick In (indicated by dashed white lines). Figure 5e and f show the \( A_{1g} \) energy maps for the 1L- and 2L-MoS\(_2\), respectively (see also Fig. S6 in Supplementary for representative Raman spectra of 1L-MoS\(_2\)). The In-covered region shows a relatively lower energy than the noncovered region, i.e., \( \Delta \omega \approx -0.3 \) and \(-1 \text{ cm}^{-1} \) for the 1L- and 2L-MoS\(_2\), respectively. These red shifts of the \( A_{1g} \) energy reflect the electron doping of the MoS\(_2\) under the In contacts. Chakraborty et. al.\(^{24}\) reported that
the $A_{1g}$ mode softens with doping at a rate of $\sim 0.2 \text{ cm}^{-1} \text{ per } 10^{12} \text{ cm}^{-2}$ for 1L-MoS$_2$, indicating that the 1L-MoS$_2$ region covered by In was doped by electrons at a density of $\sim 1.5 \times 10^{12} \text{ cm}^{-2}$. The full-width at half maximum, $\Gamma$, in Figs. 5g and h also shows consistent results. For instance, the In-covered region shows a relatively broader $\Gamma$ than the non-covered region for both 1L- and 2L-MoS$_2$, implying electron doping. The electron doping at the In/MoS$_2$ contact induces an accumulation-type contact as shown in the right panel of Fig. 5a, allowing a feasible field emission at the In/MoS$_2$ contact.

Finally, we obtained $R_cW$ for the 1L-MoS$_2$ device. In Fig. 4c, the $R_cW$ values were extracted via the TLM with three channels ($L = 0.5, 0.9, 1.4 \mu\text{m}$) in the inset of Fig. 4b. For three $V_{G-th}$ conditions of 35, 40, and 45 V, $R_cW$ decreased with decreasing $T$ in the range $250 \geq T \geq 100 \text{ K}$, representing the field emission. At $V_{G-th} = 45 \text{ V}$, $R_cW$ reached $\sim 1 \text{ k}\Omega \mu\text{m}$ at $T = 100 \text{ K}$ as the minimum value obtained from the 1L-MoS$_2$ device. This value is similar to that obtained from the 6L-MoS$_2$ device at a similar $T$ range (Fig. 2b). Interestingly, $R_cW$ increased with decreasing $T$ for $T < 100 \text{ K}$ under all $V_{G-th}$ conditions. In this region, the MoS$_2$ channel also exhibited insulating behavior (Fig. 4b) for $V_G < 60 \text{ V}$ and $T < 100 \text{ K}$. This result indicates that the increase of $R_{th}$ with decreasing $T$ in the insulating phase plays a dominant role in determining the contact resistance at $T < 100 \text{ K}$.

**Conclusion**

We have explored the contact resistance of MoS$_2$ devices with vdW In/MoS$_2$ accumulation-type contacts and revealed the contact transport mechanism for 1L- and 6L-MoS$_2$ devices. The vdW In/MoS$_2$ interface was achieved on the basis of the ‘low thermal energy’ character during an In deposition process. Whereas most metal contacts with MoS$_2$ have been prone to the FLP effect...
with a depletion contact, the In/MoS$_2$ interface provided an accumulated contact. For the 6L-MoS$_2$ device, the In/MoS$_2$ contacts provided field-emission transport as a dominant factor for $1 \times 10^{12} \leq n_e \leq 7 \times 10^{12}$ cm$^{-2}$ and $2.4 \leq T \leq 100$ K with a contact resistance of $0.6–1$ kΩ µm and a specific contact resistivity ranging from $0.8 \times 10^{-6}$ to $5 \times 10^{-6}$ Ω cm$^2$. The contact resistance was sensitive to change of sheet resistance of MoS$_2$, rather than that of the specific contact resistivity with the field-emission region. For the 1L-MoS$_2$ device, the contact transport mechanism was also sensitive to the change of the sheet resistance of MoS$_2$. On the basis of our analysis of the Raman spectra of the In/MoS$_2$ contacts, we propose that the lower contact resistance of the In/$n$L-MoS$_2$ ($n \leq 6$) devices originates from the formation of the electron accumulation-type contact at the vdW In/MoS$_2$ interface.

**Methods**

**TEM imaging** A cross-sectional TEM sample was prepared by the dual-beam focused ion beam (Helios, FEI). HR-FE-TEM (JEM-2200FSJEOL) was used for the TEM imaging at 200 kV.

**Raman spectrum** The Raman measurements were performed in a backscattering geometry at room temperature. An incident laser light with a wavelength of 514.5 nm was focused on the sample surface through an optical microscope objective lens (100×/0.9 NA). An excitation laser power was maintained less than 0.4 mW to avoid any laser-induced heating effects. Scattered light from the sample was dispersed through a monochromator with a 1200 grooves/mm grating and was collected using a thermoelectrically cooled charge-coupled device detector. For mapping measurements, Raman spectra were taken at the step of 0.5 µm over the area of $15 \times 15$ µm$^2$. 
**Electrical measurements** The electrical characterizations were performed using a physical property measurement system (PPMS, Quantum Design Inc.) with various temperatures. The $I-V_{SD}$ curves were measured by a DC bias voltage source (Yokogawa 7651) combined with a current pre-amplifier (DL 1211). The two-probe and four-probe conductance measurements for 1L-MoS$_2$ device were performed by a dc measurement setup. The two-probe and four-probe conductance measurements for 6L-MoS$_2$ device were performed by using a standard lock-in amplifier (SR830) with current preamplifier, where excitation voltage and output frequency were 30 mV and 77.77 Hz, respectively.

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**Author contributions**

M.-H.B. and J.-J.K conceived the research project. K.W. and T.T grew the bulk hBN. D.-H.C and B.-K.K fabricated the devices. D.-H.C performed the TEM analysis. B.-K.K performed the
electrical measurements and analysed the data with M.-H.B. H.K. and H.R. performed the Raman spectroscopy. M.-H.B, B.-K.K and J.-J. K wrote the manuscript. All authors discussed the results and commented on the manuscript.

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Figure captions

Figure 1| Characterization and electrical properties of In/MoS$_2$ devices. a, Optical image of a 6L-MoS$_2$/hBN device with multiple In contacts. Scale bar: 5 μm. b, Cross-sectional TEM image of the In/MoS$_2$ interface. Scale bar: 5 nm. c, Optical images before (left panel) and after (right panel) adhesive tape was used to mechanically remove the deposited In metal on a MoS$_2$ flake. Scale bar: 2 μm. d, Conductance ($G$) as a function of the back-gate voltage ($V_G$) at various temperatures. e, Current ($I$) as a function of the source–drain voltage ($V_{SD}$) for various $V_G$ at $T = 2.4$ K. f, Mobility ($\mu$) as a function of temperature ($T$) on a log scale, as obtained from 2-probe (open circles) and 4-probe (closed squares) measurements. The dashed line is a fitting line with a relation of $\mu(T) \propto T^{-\alpha}$ ($\alpha = 2.2$).

Figure 2| Contact and sheet resistance of an In/6L-MoS$_2$ device. a, Contact resistance ($R_cW$) as a function of back-gate voltage ($V_{G-th}$) for a 6L-MoS$_2$ device at various temperatures, where $V_{G-th} = V_G - V_{th}$ and $V_{th}$ is the threshold voltage. b, $R_cW$ as a function of $T$ at $V_{G-th} = 45$ V. c, $R_{sh}$ as a function of $V_{G-th}$ for various temperatures. d, $R_{sh}$ as a function of $T$ at $V_{G-th} = 45$ V.

Figure 3| Contact resistance vs. carrier density and sheet resistance. a, Solid curves and scattered triangles: $R_cW$ as a function of carrier density ($n_e$) of graphene (Gr)/4L-MoS$_2$ and In/6L-MoS$_2$ devices, respectively, at various temperatures. b, Solid curves and scattered triangles: $R_cW$ as a function of sheet resistance ($R_{sh}$) of Gr/4L-MoS$_2$ and In/6L-MoS$_2$ devices, respectively, at various temperatures. Solid, dashed, and two black dotted lines: fitting results with a relation of $R_cW \propto R_{sh}^\gamma$ ($\gamma = 7, 0.2, 0.5$, respectively). The upper and lower dotted lines were obtained with
\[ \rho_c = 5 \times 10^{-6} \, \Omega \, \text{cm}^2 \] and \[ 5 \times 10^{-7} \, \Omega \, \text{cm}^2, \] respectively. In a and b, solid and dashed ovals indicate thermionic and field-emission regimes, respectively, for the contact transport mechanism of the Gr/4L-MoS\textsubscript{2} contact at \( T = 12 \, \text{K} \).

**Figure 4** Electrical characterization and contact resistance of In/1L-MoS\textsubscript{2} device. a, Two-probe \( G-V_G \) curves at various temperatures obtained from the channel (\( L = 1 \, \mu \text{m} \)) indicated by a dashed box in the inset of b. b, Four-probe \( G-V_G \) curves at various temperatures obtained from the \( L = 1 \, \mu \text{m} \) channel. The inset of b: AFM image of a 1L-MoS\textsubscript{2} device with four In electrodes. The region outlined by a solid white line indicates the MoS\textsubscript{2} flake. Scale bar: 5 \( \mu \text{m} \). c, \( R_cW-T \) curves corresponding to various \( V_{Gth} \) conditions.

**Figure 5** Electron accumulation-type contact and Raman mapping of In/1L- and In/2L-MoS\textsubscript{2}. a, The band diagram of In and MoS\textsubscript{2} before (left panel) and after (right panel) contact. \( E_{F(m)} \) is the Fermi level of a material, \( m \). \( E_C \) and \( E_V \) are the conduction and valence bands of MoS\textsubscript{2}. b, Schematic of In/MoS\textsubscript{2} interface corresponding to the right panel of a. A, B, C, and D in the right panel of a and b indicate the current path from In to MoS\textsubscript{2} channel. c,d, Optical images of 1L- and 2L-MoS\textsubscript{2} (dashed black-boxed region) on SiO\textsubscript{2} substrates, respectively. White boxed regions: 5 nm-thick In-deposited regions. Scale bar: 5 \( \mu \text{m} \). e,f, \( A_{1g} \) energy (\( \omega \)) maps for 1L- and 2L-MoS\textsubscript{2}, respectively. g,h, Full-width at half-maximum (\( \Gamma \)) maps of \( A_{1g} \) for 1L- and 2L-MoS\textsubscript{2}, respectively.
Figure 1

(a) (b)

(c) SiO\textsubscript{2} hBN MoS\textsubscript{2} In

(d) (e)

(f) μ (cm\textsuperscript{2}V\textsuperscript{-1}s\textsuperscript{-1})

G (mS)

I (\mu A)

V (V)

2.4

T (K)

0 20 40 60 80
0.0
0.4
0.8
1.2
300
180
125
100
80
60
40
20

0 20 40 60 80
0.0
0.4
0.8
1.2
300
180
125
100
80
60
40
20

0 20 40 60 80
0.0
0.4
0.8
1.2
300
180
125
100
80
60
40
20

α = 2.2

2-probe meas.
4-probe meas.

μ (cm\textsuperscript{2}V\textsuperscript{-1}s\textsuperscript{-1})

T (K)
Figure 2

(a) Graph showing the relationship between $R_cW$ (k$\Omega$ $\mu$m) and $V_{G-th}$ (V) for different temperatures (T) in Kelvin.

(b) Graph showing the relationship between $R_cW$ (k$\Omega$ $\mu$m) and T (K) with $V_{G-th} = 45$ V.

(c) Graph showing the relationship between $R_{sh}$ (k$\Omega$) and $V_{G-th}$ (V) for different temperatures (T) in Kelvin.

(d) Graph showing the relationship between $R_{sh}$ (k$\Omega$) and T (K) with $V_{G-th} = 45$ V.
Figure 3
Figure 4
Figure 5
Supplementary for

Electron accumulation-type Ohmic contact for MoS$_2$ field-effect transistor

Bum-Kyu Kim, Dong-Hwan Choi, Hanul Kim, Kenji Watanabe, Takashi Taniguchi, Heesuk Rho, Ju-Jin Kim, and Myung-Ho Bae

Section 1. 6L-MoS$_2$ device

Figure S1. (a) Raman spectrum of 6L-MoS$_2$ device (upper panel), which was taken at the green spot in a photo-image of the lower panel. (b)-(d) upper panels: AFM images of the device. Lower panels: thickness profile along the red boxes in the corresponding upper panels ((b),(c): MoS$_2$, (d): hBN).
Figure S2. (a) 4-probe $G-V_G$ curves of 6L-MoS$_2$ device for various temperatures. (b) Normalized conductance as a function $T$ for various $V_G$. For $V_G > 15$ V corresponding to the crossover temperature between the metal and insulator characters, the down-turn curvature for $T < 70$ K was disappeared.

Figure S3. Mobility ($\mu$) as a function of $V_G$ at various temperatures for (a) two-probe and (b) four-probe measurements. The dashed lines traces the local maximum mobility for varying $T$. The mobility was obtained by a relation of $\mu = \frac{L}{W} \frac{1}{C_G} \frac{dG}{dV_G}$, where $L$ and $W$ are channel length and width, respectively, and $C_G = \left( C_{SiO_2}^{-1} + C_{hBN}^{-1} \right)^{-1}$ is the back-gate capacitance.
Section 2. 1L-MoS2 device

Figure S4. (a) Schematic of 6L-MoS2 device for the transfer length method (TLM). Here, $L_c (= 1 \mu m)$ is the length of contact electrode. The measurement scheme shows the two-probe measurement for the $L_2 = 1 \mu m$ channel. (b) Conductance as a function of $V_{G-th}$ for the four channels at $T = 210$ K. (c) Total resistance ($R_T$) as a function channel length $L$ for representative $V_{G-th}$ values at $T = 210$ K. $R_c$ and $L_T$ are the contact resistance and transfer length, respectively. $L_T$ was estimated as $\sim 0.1 \mu m$.

Figure S5. (a) AFM image of 1L-MoS2 device. (b) Thickness profile along the red line in (a).
Section 3. 1L-MoS$_2$ to study the doping effect by In contacts

Figure S6. (a) Optical photo-image of a 1L-MoS$_2$ (outlines: dashed black lines) on SiO$_2$ with indium contacts (dashed white lines). (b) and (c) Scattered points: Raman spectra obtained at white (In/1L) and black (1L) cross marks in (a). Green and blue curves are Lorentzian-fit results to estimate the energy and width of phonon modes.