The achievement of ultraclean Ohmic van der Waals (vdW) contacts at metal/transition-metal dichalcogenide (TMDC) interfaces would represent a critical step for the development of high-performance electronic and optoelectronic devices based on two-dimensional (2D) semiconductors. Herein, we report the fabrication of ultraclean vdw contacts between indium (In) and molybdenum disulfide (MoS2) and the clarification of the atomistic origins of its Ohmic-like transport properties. Atomically clean In/MoS2 vdw contacts are achieved by evaporating In with a relatively low thermal energy and subsequently cooling the substrate holder down to ~100 K by liquid nitrogen. We reveal that the high-quality In/MoS2 vdw contacts are characterized by a small interfacial charge transfer and the Ohmic-like transport based on the field-emission mechanism over a wide temperature range from 2.4 to 300 K. Accordingly, the contact resistance reaches ~600 Ωμm and ~1000 Ωμm at cryogenic temperatures for the few-layer and monolayer MoS2 cases, respectively. Density functional calculations show that the formation of large in-gap states due to the hybridization between In and MoS2 conduction band edge states is the microscopic origins of the Ohmic charge injection. We suggest that seeking a mechanism to generate strong density of in-gap states while maintaining the pristine contact geometry with marginal interfacial charge transfer could be a general strategy to simultaneously avoid Fermi-level pinning and minimize contact resistance for 2D vdw materials.

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**RESULTS AND DISCUSSION**

Characterizations of the ultraclean In/MoS$_2$ interface

We fabricated MoS$_2$ field-effect transistors (FETs) on hexagonal boron nitride (h-BN) flakes, where the h-BN flakes were deposited onto a 300-nm-thick SiO$_2$/Si substrate by mechanical exfoliation. We then transferred a few-layer MoS$_2$ flake (HQ-graphene, Inc.) onto a selected h-BN flake$^{12,20}$. For the electrical measurements, we deposited 100-nm-thick In electrodes across the MoS$_2$ channel, where the substrate holder was kept at ~100 K by flowing liquid nitrogen through it (see Fig. 1a). The substrate cooling process leads to an important result, namely a highly uniform surface morphology of In film. This contrasts strongly with the usage of a room-temperature holder that produces a segregated granular film for at least up to ~70 nm thickness, as reported in our previous work (also see Supplementary Fig. 1)$^{21}$. The upper panel of Fig. 1b shows a cross-sectional TEM image of the In/few-layered MoS$_2$ junction, which clearly shows an atomically separated interface between In and MoS$_2$ layers without any metal invasion into the MoS$_2$ layers. Whereas crystal-lattice disorders that cause defect-induced gap states and FLP typically occur during the high-temperature deposition process of evaporated metal atoms with high thermal energy$^{7,14}$, the In deposited at a relatively low thermal energy could apparently provide a clean vdW interface without disorder or defect. For instance, whereas the evaporation temperature of Au at $10^{-7}$ Torr is ~860 °C, only ~530 °C is required for the evaporation of In at the same pressure. For comparison, we prepared an Au/few-layered MoS$_2$ junction, where the substrate holder was also kept at ~100 K during Au deposition. The lower panel of Fig. 1b shows a TEM image of the Au/MoS$_2$ junction, where we observe that the invasion of Au atoms during the deposition process produces atomic defects at the first and second layers from the MoS$_2$ interface, like previous studies$^{14}$. To further characterize the quality of In/MoS$_2$ interface, we applied the Raman spectroscopy for both the pristine and In-covered MoS$_2$ regions to estimate doping effect at the metal contacts. It is known that the A$_{1g}$ phonon peak of MoS$_2$ exhibits a red shift and its width broadens with electron doping$^{22,23}$. Figure 2a, b shows the optical images of, respectively, 1L- and bilayer (2L)-MoS$_2$ (indicated by regions bounded with dashed black lines) prepared on SiO$_2$ and partially covered with 5-nm-thick In (indicated by regions bounded with dashed white lines).

Figure 2c, d shows the A$_{1g}$ energy maps for the 1L- and 2L-MoS$_2$, respectively. The In-covered region shows a relatively lower energy than the non-covered regions, i.e., $\Delta \omega \approx -0.3$ and $-1$ cm$^{-1}$ for the 1L- and 2L-MoS$_2$, respectively (see Supplementary Fig. 2 for representative Raman spectra for a 1L-MoS$_2$). In the upper panel of Supplementary Fig. 2a, we also show the $E_{2g}$ energy map of the 1L-MoS$_2$, which indicates relatively negligibly difference between the In-covered and non-covered regions. For the biaxial strain, the red shift of the $E_{2g}$ Raman mode corresponding to the in-plane vibration is more sensitive than the A$_{1g}$ mode (out-of-plane vibration)$^{24,25}$. For the in- and out-of-plane compressive strains, both modes should show blue shifts$^{26}$. In our case, however, we observed a relatively strong red shift of the A$_{1g}$ mode than that of the $E_{2g}$ mode, which has been interpreted as a doping effect$^{22}$. For instance, Chakraborty et al.$^{22}$ reported that the A$_{1g}$ mode softens with doping at a rate of ~0.2 cm$^{-1}$ per $10^{12}$ cm$^{-2}$ for 1L-MoS$_2$, whereas the $E_{2g}$ mode is relatively insensitive to the doping. We thus conclude that the 1L-MoS$_2$ region covered by In was doped by electrons at a density of ~$1.5 \times 10^{12}$ electrons cm$^{-2}$ (electron accumulation in MoS$_2$). The full-width at half-maxima, $\Gamma$, in Fig. 2e, f also show 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. This In-to-MoS$_2$ electron transfer feature will be further discussed below based on DFT calculations and shown to be another strong indication of ultraclean vdW contacts.

Charge redistribution at the In/MoS$_2$ interface

To extract the atomistic information of the In/MoS$_2$ vdW contact and contrast them with those of the Au/MoS$_2$ counterpart, we carried out DFT calculations for the vertical In/MoS$_2$ and Au/MoS$_2$ interface models and applied several analysis methods$^{27,28}$. In Fig. 3, b top panels, we show the fully optimized In/MoS$_2$ and Au/MoS$_2$ contact models, respectively (see details in Supplementary Figs. 3, 4 and Table 1). First, compared to the Au/MoS$_2$ case, we find in the optimized In/MoS$_2$ atomic structure negligible structural distortions at the rightmost metal atomic layer (see also Supplementary Fig. 3). As supported by the DFT-estimated binding energy$^{29,30}$ as well as the explicit experimental demonstration for the transferred Au electrode$^{14}$, Au is a representative metal that forms vdW-type interactions.
with MoS₂. Accordingly, the comparatively smaller In contact-induced structural perturbations consistently seen in our experiment and simulation indicate that In forms even more ideal vdW interactions with MoS₂ than Au.

To quantify this conclusion, we calculated the real-space charge density differences (Δρ) at the metal/MoS₂ interfaces according to

\[
\Delta \rho = \rho_{\text{metal/MoS}_2} - \left( \rho_{\text{MoS}_2} + \rho_{\text{metal}} \right)
\]  

and overlaid the results on the atomic structures in Fig. 3a, b bottom panels. The plane-averaged Δρ(z) for the In/MoS₂ and Au/MoS₂ contact cases are also presented in Fig. 3a, b middle panels, respectively. A positive (negative) Δρ indicates a gain (loss) in electron density, and we find stronger charge redistributions in the Au/MoS₂ contact compared with the In/MoS₂ counterpart.

Examining the distribution of Δρ between the surface metal layers and the interfacial S layer of MoS₂, we further note that due to the “push-back” effect arising from Pauli repulsion there appear charge-depleted (negative Δρ) 2D plane regions close to the metal surfaces (denoted by vertical dotted lines). Using the minimum-Δρ layers as the reference planes (z = 0), we calculated along the MoS₂ direction the position-dependent...
channels were nearly identical at 2.4 K. 

raman measurement (see Fig.2). The marginal electron transfer and the above-described estimate of $\approx 1.5 \times 10^{12}$ electrons cm$^{-2}$ is in good quantitative agreement with the density of 1L-#1 MoS$_2$ and 6L-#2 MoS$_2$ devices reached $\approx 600$ cm$^{-2}$ with respect to the metal work levels of 1L-#1 MoS$_2$ and 6L-#2 MoS$_2$ devices, respectively. As expected value for bulk MoS$_2$ ($\sim 50$ cm$^{-2}$), in both cases, the data were fitted with a relation of $\mu(T) = \mu_0 T^{-\alpha}$ with $\mu_0 = 2.2$ as shown by the dashed red line. This value is close to the expected value for bulk MoS$_2$ ($\alpha = 2.6$) with the optical phonon scattering as a dominant scattering mechanism. At room temperature, $\mu = 50$ cm$^{-2}$ V$^{-1}$ s$^{-1}$ values were obtained for both cases. However, the two-probe and four-probe $\mu$ values were saturated with decreasing temperature in the region $T < 20$ K at 1200 and 3200 cm$^{-2}$ V$^{-1}$ s$^{-1}$, respectively. The saturation behavior in low-$T$ regions is known to occur when the impurity scattering assumes a dominant role while the phonon-scattering effect is suppressed. By contrast, the mobility for two different 1L MoS$_2$ devices (1L-#1 and 1L-#2; see Supplementary Figs. 10c, 13, respectively) displayed a stability against aging at least for 40 days for $V_G < 50$ V (see Supplementary Fig. 11d).

In Fig. 4e, we show the field-effect mobility ($\mu$) of the 6L-#1 MoS$_2$ device as a function of $T$ obtained from the two-probe (open squares) and four-probe (closed squares) measurement schemes. The mobility was obtained at the local maximum location in the $\mu$-$V_G$ curves (see Supplementary Fig. 12). For $T > 100$ K, in both cases, the data were fitted with a relation of $\mu(T) = \mu_0 T^{-\alpha}$ with $\alpha = 2.2$ as shown by the dashed red line. This value is close to the expected value for bulk MoS$_2$ ($\alpha = 2.6$) with the optical phonon scattering as a dominant scattering mechanism. At room temperature, $\mu = 50$ cm$^{-2}$ V$^{-1}$ s$^{-1}$ values were obtained for both cases. However, the two-probe and four-probe $\mu$ values were saturated with decreasing temperature in the region $T < 20$ K at 1200 and 3200 cm$^{-2}$ V$^{-1}$ s$^{-1}$, respectively. The saturation behavior in low-$T$ regions is known to occur when the impurity scattering assumes a dominant role while the phonon-scattering effect is suppressed. By contrast, the mobility for two different 1L MoS$_2$ devices (1L-#1 and 1L-#2; see Supplementary Figs. 10c, 13, respectively) displayed a stability against aging at least for 40 days for $V_G < 50$ V (see Supplementary Fig. 11d).
Fig. 5 Schottky barrier height at In/MoS2 contacts. a G–V_G curves at various temperatures obtained from the L = 1 μm channel of 1L-#2 MoS2 device, indicated by a dashed box in Supplementary Fig. 13a. b Scattered points: ln(I/T^3/2) as a function of 1/k_B T with V_SD = 0.5 V from V_G = 0 V to 70 V with 5 V spacing (from bottom to top) obtained from the L = 1 μm channel. Solid lines: fitting result to obtain slope values for each V_G value. c Scattered points: slope as a function of V_SD for various V_G (5 V to 70 V with 5 V spacing from bottom to top). Solid lines: fitting result to obtain φ_{AE} at V_SD = 0 V for each V_G value. d φ_{AE} as a function of V_G for L = 1 and 1.5 μm channels. The arrow indicates the φ_{SB} (~7 meV) for the 1L-#2 MoS2 device.

Schottky barrier height at In/MoS2 contacts

We evaluated φ_{SB} at the In/MoS2 interface because this parameter plays a critical role in determining the contact resistance between a metal and a semiconductor. For this purpose, it is necessary to measure the activation energy (φ_{AE}) at the contacts in the thermionic emission region. Here, because the V_G range for the insulating region is larger than that of the 6L-#1 MoS2 device (see Fig. 4c), we used the 1L-#2 MoS2 device with the L = 1 μm channel shown in Supplementary Fig. 13a. Figure 5a shows the resulting G–V_G curves for various temperatures obtained by the two-probe measurement. In this case, the crossover V_G between the insulating and metallic regions was located at a relatively higher V_G (~65 V) than that of the 6L-#1 MoS2 device, as indicated by an arrow. The G values increased with increasing T for T < 130 K at V_G < 65 V and decreased for T > 200 K in the examined V_G range. When the thermionic emission is dominant, the current crossing a metal/2D system is described by the relation

I_d = A^* T^{3/2} \exp \left( \frac{-e \phi_{AE}}{k_B T} \right) \cdot \left( \frac{e V_G}{\eta k_B T} \right) - 1 ,

(3)

where A^* is the Richardson constant, e is the elementary charge, k_B is the Boltzmann constant, and η is the ideality factor that accounts for a lower barrier height due to image charging. Figure 5b shows ln(I/T^3/2) as a function of 1/k_B T at V_SD = 0.5 V for various V_G values from 0 V to 70 V with 5 V spacing (from bottom to top). The slopes of ln(I/T^3/2) – 1/k_B T curves are related to φ_{AE} as φ_{AE} = V_SD/η - slope. After obtaining the slopes corresponding to representative V_SD values, we plotted the slope as a function of V_SD for various V_G (5 V to 70 V with 5 V spacing from bottom to top) to obtain φ_{AE} at V_SD = 0 V, as shown in Fig. 5c. Finally, we plotted φ_{AE} vs. V_G to obtain φ_{SB} as blue squares in Fig. 5d. Near the depletion region, φ_{AE} is linearly lowered with increasing V_G when the thermally activated transport is dominant and changes its slope when the field-emission transport is accounted at the Schottky barrier. Thus, the crossover point between them occurs when the band flattens, where the value of φ_{AE} becomes equal to the value of φ_{SB}. To find V_G making the band flat, we plotted two linear blue lines on the blue squares in Fig. 5d. The two curves meet at V_G = 10 V, where the flat band is believed to form, and we estimated φ_{SB} = 7 meV at the corresponding φ_{AE} for the In/MoS2 (n = 1) contact. In addition, we also obtained a similar value for the L = 1.5 μm channel as indicated by green squares and green fit lines in Fig. 5d (see also Supplementary Fig. 13a). This value is in a similar range obtained from a Co/BN contact with a monolayer MoS2 (ref. 10). Such a low φ_{SB} at the In/MoS2 contact could allow the field-emission to play a dominant role for the transport across the In/MoS2 contact.

Contact resistance at In/MoS2 contacts

On the basis of the TLM measurements (see Supplementary Fig. 15) with multiple channels (see Fig. 4a), we extracted the contact resistance (R_C) as a function of n_e of the 6L-#1 MoS2 device at representative temperatures; the results are shown in Fig. 6a (solid squares; see also Supplementary Table 2). Here, n_e was estimated from the relation n_e = (eμR_th)^{-1}. We note that, for the consistency, the sheet resistance R_th, as well as mobility μ were obtained from the four-probe data, although R_th could have been extracted from the TLM method. The obtained contact resistance includes serial resistances of In and Ti/Au electrodes (see supplementary Fig. 16). At a given T, R_C decreased with increasing n_e. The contact
Sheet resistance vs. specific contact resistivity

We further analyzed the mechanism of charge transport across In/ MoS2 contacts with experimental data in detail. We first examined which component between \( R_n \) and \( \rho_c \) predominantly determines the contact resistance of the In/MoS2 contact. For comparison, we included in Fig. 6a other values reported in the literature; graphene(Gr)/four-layer (4L)-MoS2 contact \( ^{12} \), Au/4L-MoS2 (ref. \( ^{35} \)) and Au/in/layer MoS2 (ref. \( ^{17} \)). In the case of Gr/4L-MoS2, the graphene functions as a work-function-controllable contact material, which leads to a lower contact resistance, i.e., \( R_{\text{sh}} \approx 1 \) k\( \Omega \) \( \mu \)m at \( n_e > 4 \times 10^{12} \) cm\(^{-2} \) and \( T = 12 \) K (see the red curve in Fig. 6a). Although both the Gr-and In-contact MoS2 devices gave a similar minimum \( R_{\text{sh}} \) at cryogenic temperatures, we conclude that the transport mechanisms at the contacts rather differ from each other. For the In/MoS2 contact case, \( R_{\text{sh}} \) decreased with decreasing \( T \) in the examined \( n_e \) range, representing the field emission (or tunneling) for all examined \( T \) and \( n_e \) ranges. However, the \( R_{\text{sh}}-n_e \) curves obtained at \( T = 12 \) and 250 K for the Gr/4L-MoS2 device suggest that the left and right sides with respect to \( n_e \approx 2 \times 10^{12} \) cm\(^{-2} \) followed the thermionic and field emissions at these values respectively. At \( T = 12 \) K for 4L-MoS2 device, although the \( R_{\text{sh}} \) of \( \approx 1 \) k\( \Omega \) \( \mu \)m was relatively insensitive to the variation of \( n_e \) in the range from \( 4 \times 10^{12} \) to \( 7 \times 10^{12} \) cm\(^{-2} \), it rapidly changed from 1 k\( \Omega \) \( \mu \)m to 6 k\( \Omega \) \( \mu \)m when \( n_e \) decreased from \( 3 \times 10^{12} \) cm\(^{-2} \) to \( 1.5 \times 10^{12} \) cm\(^{-2} \). In the case of the Au/4L-MoS2 contact, on the other hand, \( R_{\text{sh}} \) was increased with decreasing \( T \) representing the thermionic emission for \( n_e < 4 \times 10^{12} \) cm\(^{-2} \). While the Au/in/layer MoS2 contact provides a relatively low-\( R_{\text{sh}} \) level for \( n_e > 1.5 \times 10^{12} \) cm\(^{-2} \) at \( T = 300 \) K, it is hard to judge the transport mechanism for the In contact because of the absence of the \( T \)-dependence of \( R_{\text{sh}} \) in the experiment. Then, for the In/6L-#1 MoS2 device, the \( R_{\text{sh}} \) was lowered with decreasing \( T \) at a given \( n_e \), for \( 1 \times 10^{12} \) cm\(^{-2} \leq n_e < 1 \times 10^{13} \) cm\(^{-2} \), representing an Ohmic-like behavior based on the field-emission mechanism in the examined \( n_e \) region. In our In/6L-MoS2 device (see Fig. 7a), \( R_{\text{sh}} \) varies in the range from 1 to 80 k\( \Omega \) when \( \rho_c \) only varies from \( 5 \times 10^{-6} \) to \( 5 \times 10^{-7} \) \( \Omega \) \( \mu \)m, as shown by two dashed lines, for \( R_{\text{sh}} \) changing from 0.6 to \( 3 \) k\( \Omega \) \( \mu \)m. This result indicates that \( R_{\text{sh}} \) plays a dominant role in determining \( R_{\text{sh}} \) in the field-emission region.

We also obtained \( R_{\text{sh}} \) from the 1L-#2 MoS2 device on a 40-nm-thick h-BN flake (see Fig. 7b and Supplementary Fig. 13 for the thickness profile). In Fig. 7b, the \( R_{\text{sh}} \) values were extracted via the TLM with three channels \( (L = 0.5, 1, 1.5 \) \( \mu \)m) as shown in Supplementary Fig. 13a. For three \( V_G \)-in, conditions of 35, 40, and 45 V, \( R_{\text{sh}} \) decreased with decreasing \( T \) in the range 250 \( \geq T \geq 100 \) K, representing the field emission. Here, \( V_{G\text{-in}} = V_G - V_t \) and \( V_{G\text{-in}} \) is a threshold voltage. At \( V_G = 45 \) V, \( R_{\text{sh}} \) reached \( \approx 1 \) k\( \Omega \) \( \mu \)m at \( T = 100 \) K as the minimum value obtained from the 1L-MoS2 device. Although this value is similar to that obtained from the 6L-MoS2 device at a similar \( T \) range (see Fig. 6a), the contact resistance for the monolayer MoS2 is higher than that of multilayer MoS2. The reason could be related to the relatively low affinity energy in the monolayer MoS2. Interestingly, \( R_{\text{sh}} \) increased with decreasing \( T \) for \( T < 100 \) K under all \( V_G \)-in conditions. In this region, the MoS2 channel also exhibited an insulating behavior in \( G \)-\( V_G \) curves for various temperatures of Fig. 5a for \( V_G < 60 \) V and \( T < 100 \) K. Thus, it indicates that the increase of \( R_{\text{sh}} \) with decreasing \( T \) in the insulating phase plays a dominant role in determining the contact resistance at \( T < 100 \) K. We note that this non-Ohmic behavior could not be improved by the contact engineering because the behavior originates from the intrinsic property of MoS2 itself. This implies that the manipulation of the metal-insulator crossover gate voltage could be crucial to get a better contact property in a mono-layer MoS2 device. In Fig. 7a (see also Supplementary Table 2), we compared the lowest achievable contact resistance as a function of \( R_{\text{sh}} \) from previous reports with

\[
R_{\text{sh}}(n_e, T) = \sqrt{R_{\text{th}}(n_e, T) \rho_c(n_e, T)},
\]

which is only valid for \( L > L_T \). Here, \( \rho_c \) is the specific contact resistivity and \( L_T = \sqrt{\rho_c/R_{\text{th}}} \) is the transfer length, which represents the average distance that charge carriers flow in a semiconductor beneath the contact before they completely transport to the electrode. Supplementary Fig. 15c shows that our device satisfied this condition with \( L_e \approx 1 \) \( \mu \)m and \( L_T \approx 0.1 \) \( \mu \)m. Equation (4) implies that \( R_{\text{sh}} \) decreases with increasing \( n_e \) because both \( R_{\text{th}} \) and \( \rho_c \) generally decrease with increasing \( n_e \). At a fixed \( n_e \), the thermionic emission charge transport mechanism across the Schottky barrier predicts that \( R_{\text{sh}} \) will increase with decreasing \( T \) because the thermionic emission will be suppressed with lowering \( T \). On the other hand, \( R_{\text{sh}} \) in our measurements decreased with decreasing \( T \), as shown by scattered red diamonds in Fig. 6b for the case measured at \( n_e = 3.4 \times 10^{12} \) cm\(^{-2} \). The contact resistance decreased from 2.3 to 0.6 k\( \Omega \) \( \mu \)m when \( T \) was decreased from room temperature to 2.4 K. This behavior, which has been reported in several previous works such as graphene/ MoS2 (ref. \( ^{12} \)) an Pd/graphene contacts \( ^{36} \), is considered as an evidence for the non-dominant role of thermionic emission for the transport across a contact. Scattered squares in Fig. 6b also show \( R_{\text{sh}} \) as a function of \( T \) at \( n_e = 3.4 \times 10^{12} \) cm\(^{-2} \), where \( R_{\text{sh}} \) decreased with decreasing \( T \) because the phonon scattering is reduced.

**Fig. 6** Contact resistance with carrier density at In/MoS2 contacts. 
\( a \) Contact resistance \( (R_{\text{sh}}, T) \) to a function of carrier density \( (n_e) \) of 6L-#1 MoS2 device at various temperatures (scattered solid squares) with other works (solid curves: graphene(Gr)/4L-MoS2, opened diamonds; Au/4L-MoS2, opened pentagons: Au/In/few-L-MoS2). \( b \) \( R_{\text{sh}} \) and sheet resistance \( (R_{\text{sh}}) \) of 6L-#1 MoS2 device as a function of \( T \) at \( n_e = 3.4 \times 10^{12} \) cm\(^{-2} \).
Atomic origins of the field emission-dominated charge transport across In/MoS$_2$ contacts

Analyzing the electronic structures of the In/MoS$_2$ and Au/MoS$_2$ interfaces obtained from DFT calculations, we finally identify the atomistic mechanisms of the experimentally observed Ohmic-like charge transport behavior. It should be noted that, while there exist in the literature several theoretical studies that examined the Schottky barriers in metal/TMD interfaces, the In/TMDC case has been rarely treated. We show the calculated band structures at the In/MoS$_2$ and Au/MoS$_2$ contacts in Fig. 8a, b, respectively, and particularly display the projected bands of Mo-4d (green circles in Fig. 8a, b), In-5p$_z$ (wine filled circles in Fig. 8a), and Au-6s (orange circles in Fig. 8b) orbitals. From the band structures, one could determine the electron $\phi_{SB}$ by measuring the energy level difference between the conduction band minimum (CBM) edge (upper solid purple line) and the Fermi level $E_F$ (dashed purple line) of metal/MoS$_2$ contacts. However, the comparison of the two bands indicate that identifying the electron $\phi_{SB}$ in the In/MoS$_2$ contact is a non-trivial matter due to the strong density of in-gap states appearing below and around the MoS$_2$ CBM region.

In summary, carrying out a combined experimental and theoretical investigation for an ultraclean vdW contact between an elemental metal In (without alloying) and semiconductor MoS$_2$, we revealed the mechanism of Ohmic charge transport across the In/MoS$_2$ vdW interface. For the single- and few-layer MoS$_2$ devices, the contact resistance decreased with decreasing temperatures for $100 \leq T \leq 300 \, \text{K}$, indicating the field-emission mechanism for the Ohmic-like contact transport. The contact resistance was sensitive to the change of sheet resistance of MoS$_2$, rather than...
that of the specific contact resistivity within the field-emission region. For the monolayer MoS$_2$ case, we achieved the contact resistance of $\sim$1 k$\Omega$$\mu$m at $T = 100$ K, which is the lowest value achieved by metal evaporations on MoS$_2$ to date. Our experimental findings were corroborated by DFT calculations, which showed that the In/MoS$_2$ contact has a weakened FLP due to the marginal interfacial charge transfer. Importantly, in spite of the weak interface dipole formation, we found that strong density of marginal interfacial charge transfer could prove to be a general strategy to seek a mechanism of introducing strong density of in-gap states while maintaining the ideal contact geometry with weak defects within the highly efficient charge injection across vdW metal contacts could be achieved via large in-gap states generated from topological defects within the sp$^2$ carbon network. We thus suggest that seeking a mechanism of introducing strong density of in-gap states while maintaining the ideal contact geometry with weak charge transfer could prove to be a general strategy to simultaneously avoid FLP and minimize contact resistance for low-dimensional vdW materials.

**METHODS**

**Device fabrication**

We fabricated MoS$_2$ FETs on h-BN flakes, where the h-BN flakes were deposited onto a 300-nm-thick SiO$_2$/Si substrate by mechanical exfoliation. We then transferred a few-layer MoS$_2$ flake (HQ-graphene, Inc.) onto a selected h-BN flake. For the electrical measurements, we deposited 100-nm-thick In electrodes across the MoS$_2$ channel, where the substrate holder was kept at $\sim$100 K by flowing liquid nitrogen through it. The substrate cooling process leads to an important result; a uniform surface morphology of In film is achieved, which contrasts strongly with the usage of a room-temperature holder that produces a segregated granular film for at least up to $\sim$70 nm thickness, as reported in our previous work (also see Supplementary Fig. 1).

**Raman spectroscopy**

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$^{-1}$ 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 $\mu$m over the area of 15 $\times$ 15 $\mu$m$^2$.

**DFT calculations**

We performed DFT calculations within the local density approximation (LDA) using the SIESTA software. The atomic cores were replaced by norm-conserving nonlocal pseudopotentials of the Troullier-Martins type, and double $\zeta$-plus-polarization-level numerical atomic orbital basis sets were employed. Geometry optimizations were performed until the Hellmann-Feynman ionic forces were below 0.02 eV Å$^{-1}$. To check the reliability of geometries and electronic structures obtained within LDA in terms of vdW interactions, we also carried out DFT calculations using the DFT-D3 (ref. 46) exchange-correlation functional. As in our earlier work, we obtained consistent results from LDA and DFT-D3 calculations (see Supplementary Figs. 3, 4 and Table 1).

**DATA AVAILABILITY**

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request. All data generated or...
REFERENCES

1. Manzeli, S., Ovchinnikov, D., Pasquier, D., Yazeyev, O. V. & Kis, A. 2D transition metal dichalcogenides. *Nat. Rev. Mater.* 2, 17033 (2017).

2. Li, C., Zhou, P. & Zhang, D. W. Devices and applications of van der Waals heterostructures. *J. Semicond.* 38, 031005 (2017).

3. Xia, F., Wang, H., Xiao, D., Dubey, M. & Ramasubramaniam, A. Two-dimensional material nanophotonics. *Nat. Photonics* 8, 899 (2014).

4. Allain, A., Kang, J., Banerjee, K. & Kis, A. Electrical contacts to two-dimensional semiconductors. *Nat. Mater.* 14, 1195 (2015).

5. Schulman, D. S., Arnold, A. J. & Das, S. Contact engineering for 2D materials and devices. *Chem. Soc. Rev.* 47, 3037 (2018).

6. Das, S., Chen, H. Y., Penumatcha, A. V. & Appenzeller, J. High performance multilayer MoS2 transistors with scandium contacts. *Nano Lett.* 13, 100 (2013).

7. Kim, C. et al. Fermi level pinning at electrical metal contacts of monolayer molybdenum disulfide. *ACS Nano* 11, 1588 (2017).

8. Yang, L. et al. Chloride molecular doping technique on 2D materials: WSe2 and MoS2. *Nano Lett.* 14, 6275–6280 (2014).

9. Wang, J. et al. High mobility MoS2 transistor with low Schottky barrier contact by using atomic thick h-BN as a tunneling layer. *Adv. Mater.* 28, 8302 (2016).

10. Cui, X. et al. Low-temperature Ohmic contact to monolayer MoS2 by van der Waals bonded Co/h-BN electrodes. *Nano Lett.* 17, 4781 (2017).

11. Leong, W. S. et al. Low resistance metal contacts to MoS2 devices with nickel-etched-graphene electrodes. *ACS Nano* 9, 869 (2015).

12. Cui, X. et al. Multi-terminal transport measurements of MoS2 using a van der Waals heterostructure device platform. *Nat. Nanotechnol.* 10, 534 (2015).

13. Kappera, R. et al. Phase-engineered low-resistance contacts for ultrathin MoS2 transistors. *Nat. Mater.* 13, 1128 (2014).

14. Liu, Y. et al. Approaching the Schottky-Mott limit in van der Waals metal-semiconductor junctions. *Nature* 557, 696 (2018).

15. Wang, J. et al. Low-power complementary inverter with negative capacitance 2D semiconductor transistors. *Adv. Funct. Mater.* 30, 2003859 (2020).

16. Kong, L. et al. Doping-free complementary WSe2 circuit via van der Waals metal integration. *Nat. Commun.* 11, 1866 (2020).

17. Wang, Y. et al. Van der Waals contacts between three-dimensional metals and two-dimensional semiconductors. *Nature* 568, 70 (2019).

18. Kim, B.-K. et al. Genuine Ohmic van der Waals contact between indium and MoS2. Preprint at https://arxiv.org/abs/1904.10295 (2019).

19. Laur, C. S. et al. Quantum transport in two-dimensional WSe2 with high-efficiency carrier injection through indium alloy contacts. *ACS Nano* 14, 13700–13708 (2020).

20. Dean, C. R. et al. Boron nitride substrates for high-quality graphene electronics. *Nat. Nanotechnol.* 5, 722 (2010).

21. Choi, D.-H. et al. Van-der-Waals-gap tunneling spectroscopy for single-wall carbon nanotubes. *Carbon* 113, 237 (2017).

22. Chakraborty, B. et al. Symmetry-dependent phonon renormalization in monolayer MoS2 transistor. *Phys. Rev. B* 85, 161403 (2012).

23. Sovranshi, D. et al. Nature of carrier injection in metal/2D-semiconductor interface and its implications for the limits of contact resistance. *Phys. Rev. B* 96, 205423 (2017).

24. Lloyd, D. et al. Band gap engineering with ultralarge biaxial strains in suspended monolayer MoS2. *Nano Lett.* 16, 5836 (2016).

25. Li, H. et al. Optoelectronic crystal of artificial atoms in strain-textured molybdenum disulphide. *Nat. Commun.* 6, 7381 (2015).

26. Hui, Y. Y. et al. Exceptional tunability of band energy in a compressively strained trilayer MoS2 sheet. *Nano Lett.* 7, 7126 (2013).

27. Kim, Y.-H. & Kim, H. S. Anomalous length scaling of carbon nanotube-metal contact resistance: an ab initio study. *Appl. Phys. Lett.* 100, 213113 (2012).

28. Kim, H. S., Lee, G. I., Kim, H. S., Kang, J. K. & Kim, Y.-H. Intrinsically low-resistance metal/dichalcogenide contacts in monolayer MoS2 transistors. *Nano Lett.* 14, 1714 (2014).

29. Guo, Y., Liu, D. & Robertson, J. 3D behavior of Schottky barriers of 2D transition-metal dichalcogenide semiconductors. *Appl. Mater. Interfaces* 7, 25709 (2015).

30. Cui, X. et al. Interfaceal properties of monolayer and bilayer MoS2 contacts with metals: beyond the energy band calculations. *Sci. Rep.* 6, 21786 (2016).

31. Kang, J., Liu, W., Sarkar, D., Jena, D. & Banerjee, K. Computational study of metal contacts to monolayer transition-metal dichalcogenide semiconductors. *Phys. Rev. X* 4, 031005 (2014).

32. Leonard, F. & Talin, A. A. Electrical contacts to one- and two-dimensional nanomaterials. *Nat. Nanotechnol.* 6, 773 (2011).

33. Ceperley, D. M. & Alder, B. J. Ground state of the electron gas by a stochastic method. *Phys. Rev. Lett.* 45, 566 (1980).

34. Soler, J. M. et al. The SIESTA method for ab initio order-N materials simulation. *J. Phys. Condens. Matter* 14, 2745 (2002).

35. Troullier, N. & Martins, J. L. Efficient pseudopotentials for plane-wave calculations. *Phys. Rev. B* 43, 1993 (1991).

36. Grimme, S., Antony, J., Ehrlich, S. & Krieg, H. A consistent and accurate ab initio parametrization of density functional dispersion correction (DFT-D) for the 94 elements H-Pu. *J. Chem. Phys.* 132, 154104 (2010).

37. Lee, J. et al. Origin and control of polycrystalline alignment on carbon nanotubes and graphene nanoribbons. *Adv. Funct. Mater.* 28, 1706970 (2018).

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AUTHOR CONTRIBUTIONS

M.-H.B., J.-J.K. and Y.-H.K. conceived the research project. K.W. and T.T. grew the bulk h-BN. 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 analyzed the data with M.-H.B. H.K. and H.R. performed the Raman spectroscopy. T.-H.K. and Y.-H.K. performed the DFT calculations. M.-H.B., B.-K.K., T.-H.K., Y.-H.K., and J.-J.K. wrote the manuscript. All authors discussed the results and commented on the manuscript. B.-K.K., T.-H.K., and M.-H.B. contributed equally to this work.

COMPETING INTERESTS

The authors declare no competing interests.

ADDITIONAL INFORMATION

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