Indium-contacted van der Waals gap tunneling spectroscopy for van der Waals layered materials

Dong-Hwan Choi¹ 2, Kyung-Ah Min¹, Suklyun Hong³, Bum-Kyu Kim², Myung-Ho Bae² 4 & Ju-Jin Kim¹ ²

The electrical phase transition in van der Waals (vdW) layered materials such as transition-metal dichalcogenides and Bi₂Sr₂CaCu₂O₈₊ₓ (Bi-2212) high-temperature superconductor has been explored using various techniques, including scanning tunneling and photoemission spectroscopies, and measurements of electrical resistance as a function of temperature. In this study, we develop one useful method to elucidate the electrical phases in vdW layered materials: indium (In)-contacted vdW tunneling spectroscopy for 1T-TaS₂, Bi-2212 and 2H-MoS₂. We utilized the vdW gap formed at an In/vdW material interface as a tunnel barrier for tunneling spectroscopy. For strongly correlated electron systems such as 1T-TaS₂ and Bi-2212, pronounced gap features corresponding to the Mott and superconducting gaps were respectively observed at T = 4 K. We observed a gate dependence of the amplitude of the superconducting gap, which has potential applications in a gate-tunable superconducting device with a SiO₂/Si substrate. For In/10 nm-thick 2H-MoS₂ devices, differential conductance shoulders at bias voltages of approximately ± 0.45 V were observed, which were attributed to the semiconducting gap. These results show that In-contacted vdW gap tunneling spectroscopy in a fashion of field-effect transistor provides feasible and reliable ways to investigate electronic structures of vdW materials.

Van der Waals (vdW) layered materials such as two-dimensional (2D) transition-metal dichalcogenides (TMDCs) and Bi₂Sr₂CaCu₂O₈₊ₓ (Bi-2212) have shown various electronic phases that emerge from many-body features such as a charge density wave (CDW) or superconductivity, depending on the temperature and carrier density¹–⁹. The vdW interface between dissimilar vdW materials have allowed to investigate the electronic structures of such strongly correlated electron systems. For instance, transport spectroscopy in TMDC/Bi-2212 vdW junctions has revealed gap natures due to the many-body features via the formation of a metal/superconductor proximity junction with a low vdW contact resistance¹⁰,¹¹. For tunneling spectroscopy, on the other hand, a graphite/Bi-2212 interface provided a vdW gap tunnel junction, enabling tunneling spectroscopy for the Bi-2212 superconductor¹¹,¹². However, it could be hard to apply the graphite to TMDCs for vdW tunneling spectroscopy because graphene/TMDC contacts have been used to form an Ohmic contact¹³. Thus, for feasible and reliable tunneling spectroscopy for vdW materials, it is crucial to seek a material to form a vdW tunneling gap with any vdW materials. With this purpose, we focus on indium (In) metal in this study.

For evaporated-metal/TMDC contacts, only indium (In) metal has shown vdW contact for TMDCs¹⁴,¹⁵, owing to its low evaporation temperature of ~ 500 °C. In this case, the In vdW contact provides an Ohmic contact for a few-layer TMDCs. However, the vdW contact at a cryogenic temperature could provide a vacuum tunneling gap with a high contact resistance that makes the flow of current sensitive to the electronic DOS at the interface. Indeed, vdW gap tunneling spectroscopy based on a field-effect transistor (FET) design with carbon nanotubes (CNTs) with In metal contacts was demonstrated, recently¹⁶,¹⁷. In this previous work, the local conductance peaks observed in the conductance vs bias voltage plot were attributed from the van Hove singularities corresponding to the sub-band structures of semiconducting and metallic CNTs.

In the present study, we apply a type of FET with In contacts for various vdW layered materials (i.e., 1T-TaS₂, Bi-2212, and 2H-MoS₂) to demonstrate that In-contacted vdW gap tunneling spectroscopy is a feasible method to investigate the electrical DOS of vdW layered materials. For the experiments with 1T-TaS₂, the zero-bias resistivity showed a sudden increase at T ~ 180 K as the temperature was lowered. At T = 4 K, a plot of the differential

1Department of Physics, Jeonbuk National University, Jeonju 54896, Republic of Korea. 2Korea Research Institute of Standards and Science, Daejeon 34113, Republic of Korea. 3Department of Physics and Graphene Research Institute, Sejong University, Seoul 05006, Korea. 4Department of Nano Science, University of Science and Technology, Daejeon 34113, Republic of Korea. *email: mhbae@kriss.re.kr; jujinkim@chonbuk.ac.kr
conductance for various bias voltages revealed the emergence of an energy gap, i.e., the Mott gap edge, which has been observed in the same material only using scanning tunneling and photoemission spectroscopies. The In/Bi-2212 junction also showed a superconducting gap of ~58 meV at T=4 K. The gap feature slightly decreased with increasing gate voltage, which indicates that the high-temperature superconductivity could be controlled by electric fields. Finally, for ~10 nm-thick MoS2 FETs, we observed gap features at energy levels of ~0.9 eV in differential conductance vs bias voltage curves recorded at T=4 K, corresponding to a semiconducting bandgap. The formation of the tunneling barrier with a high contact resistance for ~10 nm-thick MoS2 is inconsistent with the Ohmic contacts for few-layer (thickness ≤ 4 nm) MoS2 flakes with In contacts, which might be related to the location of the Fermi level of an In electrode with respect to the bandgap, depending on the thickness of the MoS2 layer. The naturally formed vdW tunnel gap without any artificial insulating barrier is very robust under varying temperature. Our work provides simple and reliable identification of electronic DOS using the simple FET geometry for the vdW materials without sophisticated tools such as scanning tunneling microscope.

**Measurements and results**

**Experiments.** Single crystals of 1T-TaS2 and Bi-2212 were grown by the usual iodine transport method and solid-state-reaction methods, respectively. A 2H-MoS2 single crystal was commercially purchased (HQ Graphene). We fabricated vdW material-based FETs with In contacts for TaS2, Bi-2212, and MoS2, by carrying out several microfabrication processes. The vdW material flakes on a 500 nm-thick SiO2/Si substrate were prepared via mechanical exfoliation from the vdW materials. We deposited 100 nm-thick In electrodes onto a multilayer flake using traditional electron-beam lithography and thermal deposition processes. To investigate the quality of the In/vdW material, we collected cross-sectional transmission electron microscopy (TEM) image of an In/few-layered MoS2 junction (Fig. 1a), where the thermally deposited In did not show invasion into the MoS2 layer, resulting in a well-defined vdW gap. Atomic structures of interfaces between In and two vdW materials of 2H-MoS2 and 1T-TaS2 with vdW gaps were prepared by the density functional theory (DFT) calculations as shown in Fig. 1b,c, respectively. The schematic atomic structure for In/Bi-2212 is also plotted in Fig. 1d. Figure 1e shows a schematic of a completed device for the vdW tunneling spectroscopy experiment (Fig. 1f), where the highly doped Si substrate serves as a back-gate electrode. For basic electrical characterizations of the three vdW mate-
T < 20 K (Fig. 3a), where conductance peaks corresponding to the gap edges are clearly observed at \(sd \approx \pm 0.2\) V, as shown in Fig. 3a (see also Fig. 2c). This result indicates that the Mott gap was fully developed at \(T = 10\) K. For \(T < 300\) K, a relatively flat current region is observed near zero bias, which corresponds to the conductance dip region at the same \(V_{sd}\) (indicated by two arrows). In the \(I-V_{sd}\) curve corresponding to \(V_g = 30\) V in Fig. 2c, a relatively flat current region is observed near zero bias, which corresponds to the conductance dip region at the same \(V_g\) value in Fig. 2b. The \(\frac{dI}{dV_{sd}}-V_{sd}\) curve corresponding to \(V_g = 30\) V shows a clear gap feature indicated by the bidirectional arrow, which was also observed at \(V_g = -30\) V in Fig. 2c. We speculated that the observed gap is related to the Mott gap (\(\Delta_{Mott}\)), exhibiting a gap size of \(\sim 0.4\) eV. The Mott transition in multilayer 1T-TaS\(_2\) has been previously shown to be developed in the temperature range \(180 \leq T \leq 210\) K, accompanied by a transition from the nearly-commensurate CDW (NCCDW) phase to the commensurate CDW (CCDW) phase, as revealed by scanning tunneling spectroscopy and photoelectron spectroscopy. A recent study based on resistance measurements as a function of \(T\) conducted by using a gate-controlled Li\(^+\)-ion intercalation method showed that the CCDW phase changed to the NCCDW phase with increasing gate voltage at \(T = 10\) K. In our case, the width of the conductance dip was nearly constant for \(V_g\) values spanning 60 V, possibly because of relatively less change in the carrier density in our gating method with a 500 nm-thick dielectric layer. In addition, in Fig. 2d, we displayed already reported \(\frac{dI}{dV_{sd}}-V_{sd}\) curves obtained by conventional scanning tunneling spectroscopy with the same crystal used in this study. Both of them showed a similar \(\Delta_{Mott}\) size of \(\sim 0.4\) eV, thus we believe that the contacted vdW gap tunneling spectroscopy provides a credible method to study the electronic states in vdW materials.

To investigate the phase transition, we obtained \(\frac{dI}{dV_{sd}}-V_{sd}\) curves at \(V_g = 30\) V over the temperature range \(4 \leq T \leq 210\) K, as shown in Fig. 3a, where the curves are vertically shifted as much as 20 nS for clarity. The pronounced gap feature at \(T = 4.2\) K was smeared with increasing \(T\) up to \(T \sim 140\) K and became featureless at \(T \geq 180\) K, whereas the conductance dip near zero bias was still observed. Figure 3b shows the zero-bias resistivity as a function of \(T\), as extracted from Fig. 3a at \(V_{sd} = 0\) V. A sudden increase is observed at \(T \sim 180\) K (indicated by an arrow) with decreasing \(T\). This behavior was found to be consistent with previous observations of the Mott transition accompanied by the phase transition from the NCCDW phase to the CCDW phase near this temperature, \(T_{CCDW\rightarrow Mott}\). In that Mott transition, a gap was not fully opened in the investigated \(T\) region, resulting in a so-called a pseudogap structure. In our case, such behavior was observed in the region indicated by a dashed bidirectional arrow in Fig. 3b, where the zero-bias resistivity monotonically increases with decreasing \(T\). The \(\frac{dI}{dV_{sd}}-V_{sd}\) curves corresponding to \(80 \leq T \leq 180\) K in Fig. 3a show pseudogap-hump structures at \(V_{sd} \sim \pm 0.3\) V with a finite zero-bias conductance, representing a small but finite density of state at the Fermi energy. Importantly, at \(T \leq 60\) K, the resistivity increases substantially faster with decreasing \(T\) (see Fig. 3b) and the zero-bias conductance finally decreases to nearly zero at \(T < 20\) K (Fig. 3a), where conductance peaks corresponding to the gap edges are clearly observed at \(V_{sd} \sim \pm 0.2\) V, as shown in Fig. 3a (see also Fig. 2c). This result indicates that the Mott gap was fully developed at \(T \leq 20\) K. To test the reproducibility of these results, we also fabricated two additional 1T-TaS\(_2\) devices with a thickness similar to that of the first 1T-TaS\(_2\) device, and the curves for each device showed a clear conductance peak at one polar \(V_{sd}\) of \(\sim 0.2\) V and \(0.18\) V at \(T = 4\) K, respectively, as shown in Fig. 3c (vertical arrows). Asymmetric gap features have been frequently observed in tunneling spectroscopy results for two electrodes (tip and metal contacts) with substantially different contact resistance levels (see Fig. 2d).

| Materials | 1T-TaS\(_2\) | Bi-2212 | 2H-MoS\(_2\) (thickness: 6 nm) |
|-----------|-------------|--------|-----------------------------|
| Carrier type and \(n_\text{H}\) | For 4 < \(T < 200\) K Hole \((0.4-5) \times 10^{19} \text{ cm}^{-3}\) | For \(T_c < T < 300\) K Hole \((3-6) \times 10^{11} \text{ cm}^{-3}\) | For 2 < \(T < 300\) K Electron \((1-2) \times 10^{11} \text{ cm}^{-2}\) |
| \(n_\text{H}\) (cm\(^2\) V\(^{-1}\) S\(^{-}\)) | For 1 < \(T < 10\) K 1-10 | For 3.2-7 nm thicknesses 3-11 | For 2 < \(T < 300\) K 3000-20 |

Table 1. Representative carrier type, carrier density (\(n_\text{H}\)) and Hall mobility (\(\mu_\text{H}\)) for three vdW materials.
vdW gap tunneling spectroscopy for Bi-2212. The as-grown Bi-2212 crystal used in the present study exhibits a slightly hole-overdoped character; the superconducting transition temperature ($T_c$) measured from other crystals obtained from the same batch was ~88 K. The upper panel of Fig. 4a shows an AFM image of a Bi-2212 device with $L \approx 0.4 \mu$m and $W \approx 0.4 \mu$m. The lower panel of Fig. 4a shows a height profile along the dotted line depicted in the upper panel of Fig. 4a, indicating that the thickness of the Bi-2212 is ~70 nm. Figure 4b shows $dI/dV_{sd}$ as a function of $V_{sd}$ at $V_g = -40$ V for various temperatures, where the curves are vertically shifted as much as 50 nS. At $T = 4$ K, the gap feature was observed at $V_{p^+}$ ~ 58 and $V_{p^-}$ ~ 50 mV, which are assigned as $V_{p^-}$ and $V_{p^+}$, respectively, as indicated by two red arrows in the figure. The observed gap sizes are consistent with previous observations of the superconducting gap energy, $\Delta_{1}$. The peak signature at $V_{p^+}$ is relatively weak compared with that at $V_{p^-}$, similar to the TaS$_2$ case. The $V_{p^-}$ value corresponding to the conductance peak decreases with increasing $T$, as indicated by the dashed green line, and is smeared out near the $T_c$ at $T = 80$ K. Figure 4d shows $\Delta(T)/e$ based on the expression:

$$\Delta(T) = \Delta_0 \tanh \left( \frac{\pi a}{b} \left( \frac{T_c}{T} - 1 \right) \right)$$

where $e$ is the elementary charge; $\Delta_0 = 58$ meV, $a = 2.14$, and $b = 4/3$ for a weak-coupling 2D d-wave superconductor; and $T_c = 88$ K. Although we lack exact information about the $T_c$ at $V_g = -40$ V, the data qualitatively follows Eq. (1). Thus, we conclude that the gap feature originates from the superconducting gap. We also measured $dI/dV_{sd}$ as a function of $V_{sd}$ at $V_g = -30$ V as $T$ was varied (Fig. 4c). As depicted by the dashed green line, the superconducting gap energy decreases with increasing $T$. However, we note that the gap feature relatively weakens at $V_g = -30$ V compared with that at $V_g = -40$ V. For instance, at $V_g = -30$ V, $V_{p^-}$ decreases to approximately ~46 mV at $T = 4$ K, accompanied by a reduction of the conductance peak height. The conductance peak
corresponding to the superconducting gap edge at the positive $V_{sd}$ region ($V_{p+}$) even disappears at $T = 4$ K. For comparison, we added $|V_{p-}|$ as a function of $T$ obtained at $V_g = -30$ V in Fig. 4d. Recently, Liao et al. reported that the superconductor–insulator transition in Bi-2212 could be achieved using a back-gate method with a solid ionic conductor, where the carrier density was modulated by intercalated Li+ ions. This result may indicate that the superconductivity of Bi-2212 can also be manipulated by a back-gate field with a SiO2/Si substrate design as our case, although further study is needed to quantitatively demonstrate the feasibility of this concept.

vdW gap tunneling spectroscopy for 2H-MoS2. The inset of Fig. 5a shows an AFM image of the MoS2 FET (MS1) with a thickness of ~10 nm. The MS1 device showed a traditional $n$-type transfer curve for $V_{sd} = \pm 0.5$ V at $T = 4$ K (Fig. 5a), where the $I$–$V_g$ curves show an asymmetric behavior depending on the polarity of $V_{sd}$. Figure 5b shows a $dI/dV_{sd}$ map as a function of $V_{sd}$ and $V_g$. In Schottky FETs, the zero-conductance region observed when $V_{sd}$ is swept in a depletion state decreases with positively increasing $V_g$ for an $n$-type device because the width of the Schottky barrier decreases with positively increasing $V_g$. Although the conductance map in Fig. 5b appears to show such behavior for $V_g < 30$ V (region i), a robust zero-conductance region that is independent of the change in $V_g$ was observed at $-0.45 \leq V_{sd} \leq 0.45$ V (see dashed yellow lines) in the $V_g$ region labeled as region ii. Figure 5c shows $I$ and $dI/dV_{sd}$ as functions of $V_{sd}$ at $V_g = 40$ V in region ii. Conductance shoulders, indicated by the two vertical dashed lines, are separated from each other by an energy scale of ~0.96 eV, which is close to the interval bandgap of ~1.2 eV expected for multilayer MoS2.

To confirm the consistency of the vdW gap tunneling spectroscopy, we fabricated another ~10 nm-thick MoS2 device (MS2; inset of Fig. 5d). The overall behaviors of the electrical properties of the MS2 device, such as the $n$-type behavior and the robust zero-conductance region (region ii) in Fig. 5d,e, respectively, show similar trends as those of the MS1 device. The conductance shoulders in Fig. 5f with the two vertical dashed lines provide an energy scale of ~0.92 eV at $V_g = 64$ V in region ii, which is also similar to that of the MS1 device.

To determine the origin of the robust zero-conductance region, we considered the possibility of tunnel barriers with a high contact resistance due to the vdW gap between the MoS2 and In electrodes. Figure 6a,b show the band structures of MoS2 and In with vdW gap tunnel barriers at the MoS2/In interfaces (vertical gray bars)
Figure 4. vdW gap tunneling spectroscopy for Bi-2212. (a) Upper panel: AFM image of a Bi-2212 device. Scale bar: 1 μm. Lower panel: height profile along the dashed black line in the upper panel. (b), (c) $dI/dV_{sd}$ as a function of $V_{sd}$ for various temperatures, as obtained at $V_g = -40$ and $-30$ V, respectively. Dashed green lines follow the gap edges as $T$ is varied. (d) Closed squares and circles: $|V_p|$ as a function of $T$ for $V_g = -40$ and $-30$ V, respectively. Dashed curve: a plot based on Eq. (1) for comparison.

for representative $V_g$ regions labeled as i and ii in Fig. 5b, respectively. For simplicity, we only considered the MS1 device in Fig. 5a–c. The upper and lower solid black curves correspond to the conduction-band (CB) and valence-band (VB) edges of MoS$_2$, respectively. The light-blue region under the VB edge indicates the states occupied by electrons. For $V_g = -20$ V, the MoS$_2$ band was found to be shifted upward, whereas the band edges were fixed at the junction interfaces, where the left electrode was grounded. $E_{F(In)}$ was located within the bandgap ($E_g$) without $V_{sd}$, as shown in the left panel of Fig. 6a (horizontal red line); thus, a sufficiently high $V_{sd}$ is needed to overcome the $E_g$. The middle and right panels of Fig. 6a show $V_{sd}$ conditions in which the $E_{F(In)}$ of the right electrode reaches the CB and VB edges, respectively. Nevertheless, electrons do not flow to the edges of the MoS$_2$ because they experience a large Schottky barrier width. $V_{sd}$ values greater than those corresponding to the band edges are thus needed to make a narrower Schottky barrier for the flow of electrons. With increasing $V_g$, the bands for the MoS$_2$ bend downward, leading to a relatively narrow Schottky barrier. Thus, the $\pm V_{sd}$ that allow the current to flow decrease with increasing $V_g$, corresponding to region i in Fig. 5b. In region ii in Fig. 5b, the interval between $\pm V_{sd}$ locations for the finite conductance edges were found to change only slightly as $V_g$ was varied. The band diagrams corresponding to region ii are plotted in Fig. 6b. In this region, the MoS$_2$ electronic bands were observed to be substantially bent downward for $V_g = -40$ V. Here, the alignment of $E_{F(In)}$ with the two band edges with proper $V_{sd}$ values of $-V_g$ and $+V_g$ enabled the electrons to tunnel between the electrode and MoS$_2$, under the assumption that the Schottky barrier widths were sufficiently narrow to allow the tunnel event, as indicated by horizontal arrows in the middle and right panels in Fig. 6b. The alignment resulted in no variation of the interval between $-V_g$ and $+V_g$ values, as indicated by the two dashed parallel lines in Fig. 5b. We note that the electrostatic band bending also occurs because of the vdW tunnel junction with a finite $V_{sd}$ which leads to an additional band bending in a lower direction with the positive $V_{sd}$.

The observed tunnel behavior with high contact resistances at In-contacted devices with 10 nm-thick ($n \approx 15$, where $n$ is the number of MoS$_2$ layers) MoS$_2$ layers appears to be inconsistent with the behavior of In/MoS$_2$ devices with MoS$_2$ thicknesses $\leq 4$ nm ($n \leq 6$), which exhibit Ohmic behavior$^{13}$. For this reason, we considered the location of $E_{F(In)}$. Figure 5c,f show that the location of zero bias is nearly midway between the two vertical dashed lines, which implies that $E_{F(In)}$ for $n \approx 15$ MoS$_2$ is located near the midgap, indicating non-Ohmic contact. However, in the case of few-layer MoS$_2$, $E_{F(In)}$ is located just below the CB edge, resulting in Ohmic contact under the tunneling (or field-emission) mechanism through the vdW gap$^{15}$. In this sense, performing vdW gap tunneling spectroscopy for $n \leq 6$ is not possible, although the bandgap drastically increases from $\sim 1.4$ to $\sim 1.9$ eV when $n$ is changed from 6 to 1$^2$. Additional experimental and theoretical studies are needed to understand the
Figure 5. vdW gap tunneling spectroscopy for 2H-MoS$_2$. (a), (d) Current vs. back-gate voltage ($I$–$V_g$) curves at $V_{sd} = \pm 0.5$ V and $T = 4$ K for MS1 and MS2, respectively. Insets of (a), (d): AFM images of MS1 (scale bar: 1 μm) and MS2 (scale bar: 2 μm), respectively. The thickness of the MoS$_2$ flakes in both cases was ~10 nm, corresponding to ~14 MoS$_2$ layers. (b), (e) Differential conductance ($d$/$dV_{sd}$) as a function of $V_{sd}$ and $V_g$ of MS1 and MS2, respectively. The zero-conductance region on the $V_{sd}$ axis became smaller with increasing $V_g$ in region “i” and did not change in region “ii.” The two dashed lines indicate the edges of the bandgap. (c), (f) $d$/$dV_{sd}$ and $I$ as a function of $V_{sd}$ at $V_g = 40$ and 64 V for MS1 and MS2, respectively. The left and right vertical dashed lines located at the conductance shoulders correspond to the edges of the conduction and valence bands, respectively.

(a) $V_g$: region i

(b) $V_g$: region ii

Figure 6. Band diagrams in MoS$_2$ device with various $V_g$ and $V_{sd}$ values. (a), (b) Band diagrams with zero and finite $V_{sd}$ conditions in $V_g$ regions corresponding to regions “i” and “ii” depicted in Fig. 5b,e, respectively. $E_g$ denotes the Fermi energy of In electrodes. Dark- and light-blue regions indicate bands filled by electrons of In and MoS$_2$, respectively. The upper and lower solid lines in the MoS$_2$ region represent the edges of the conduction and valence bands, respectively. $E_g$ is the energy gap of MoS$_2$. Solid red lines in the In region indicate Fermi levels depending on $V_{sd}$ conditions. Vertical red arrows depict the direction of the changing Fermi levels with finite $V_{sd}$ conditions from the zero-bias condition (dashed red lines). Horizontal red arrows represent the current flow directions via tunneling.
current flowing between In and various-thickness MoS₂ to know the limit of In-contacted vdW gap tunneling spectroscopy for MoS₂.

**Conclusions**

We carried out In-contacted vdW gap tunneling spectroscopy for 1T-TaS₂, Bi-2212, and 2H-MoS₂ using an FET geometry. We clearly observed the Mott gap (~ 0.4 eV), superconducting gap (~ 58 meV), and semiconducting bandgap (~ 0.9 eV) of 1T-TaS₂, Bi-2212, and MoS₂, respectively, by analyzing conductance curves as a function of V_G at T = 4 K. Thus, we propose that vdW gap tunneling spectroscopy provides a feasible method to reveal the electronic band structure of inert vdW layered 2D materials. For semiconductor vdW materials of MoS₂, we found that In-contacted vdW gap tunneling spectroscopy is applicable for only bulk MoS₂ (n ~ 15), which could be related to the location of the Fermi level of In with respect to the midgap of MoS₂. This reflects that the relative location of the Fermi level of In with respect to the midgap of vdW material may reveal the limitations of In-contacted vdW gap tunneling spectroscopy. For non-semiconductor vdW materials of 1T-TaS₂ and Bi-2212 with thickness of tens of nanometers, we confirmed the In-contacted vdW gap tunneling spectroscopy is applicable while we need further study for the validity for a few layers.

**Methods**

**Samples.** We fabricated electrical devices with In contacts for TaS₂, Bi-2212, and MoS₂ on 500 nm-thick SiO₂/Si substrates by conventional microfabrication processes. We deposited 100 nm-thick In electrodes onto a multilayer flake in a thermal evaporator at a vacuum of 3 × 10⁻⁶ Torr. The substrate was attached to a stage, which was cooled to 100 K using liquid N₂ during the In deposition process. The low temperature of the sample stage ensured a homogeneously deposited In layer with a uniform thickness and without grain boundaries.

**Measurements.** We performed electrical measurements using a two-probe configuration in a closed-cycle refrigerator with a base temperature of 4 K. Bias voltages were applied by Keithley 213 quad voltage source and current was measured by a current amplifier (Ithaco 1211, DL).

**Computations.** DFT calculations for Fig. 1b,c are carried out within generalized gradient approximation (GGA) for exchange–correlation (xc) functionals, implemented in the Vienna ab initio simulation package (VASP).
25. Bae, M.-H., Park, J.-H., Choi, J.-H., Lee, H.-J. & Park, K.-S. Pseudogap behavior revealed in interlayer tunneling in overdoped Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$. *Phys. Rev. B* **77**, 094519 (2008).

26. Kordyuk, A. A. Pseudogap from ARPES experiment: Three gaps in cuprates and topological superconductivity. *Low Temp. Phys.*** **41**, 319 (2015).

27. Hashimoto, M., Vishik, I. M., He, R.-H., Devereaux, T. P. & Shen, Z.-X. Energy gaps in high-transition-temperature cuprate superconductors. *Nat. Phys.* **10**, 483 (2014).

28. Prozorov, R. & Giannetta, R. W. Magnetic penetration depth in unconventional superconductors. *Supercond. Sci. Technol.* **19**, R41 (2006).

29. Mak, K. F., Lee, C., Hone, J., Shan, J. & Heinz, T. F. Atomically thin MoS$_2$: A new direct-gap semiconductor. *Phys. Rev. Lett.* **105**, 136805 (2010).

30. Kohn, W. & Sham, L. J. Self-consistent equations including exchange and correlation effects. *Phys. Rev.* **140**, A1133 (1965).

31. Perdew, J. P., Burke, K. & Ernzerhof, M. Generalized gradient approximation made simple. *Phys. Rev. Lett.* **77**, 3865 (1996).

32. Kress, G. & Furthmüller, J. Efficient iterative schemes for ab initio total-energy calculations using a plane-wave basis set. *Phys. Rev. B* **54**, 11169 (1996).

**Acknowledgements**

This research was supported by the Korea Research Institute of Standards and Science (KRISS-2021-GP2021-0001). This work was also partly supported by the National Research Foundation of Korea (NRF) (Grant Nos. SRC2016R1A5A1008184, 2018R1A2A1A05078440, 2018K1A4A3A01064272, 2020R1F1A1075216, 2021R1A2C3012612, 2021R1A4A1031900).

**Author contributions**

Supervision: M.-H. B and J.-J.K. Device and chip design: D.-H.C. and M.-H.B. Fabrication: D.-H.C. Measurements and analyses: D.-H.C. Measurements and analysis for carrier concentration and Hall mobility of MoS$_2$: B.-K.K. DFT calculations: K.-A.M and S.H. Manuscript composition: All authors.

**Competing interests**

The authors declare no competing interests.

**Additional information**

**Supplementary Information** The online version contains supplementary material available at https://doi.org/10.1038/s41598-021-97110-z.

**Correspondence** and requests for materials should be addressed to M.-H.B. or J.-J.K.

**Reprints and permissions information** is available at www.nature.com/reprints.

**Publisher’s note** Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

**Open Access** This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are included in the article’s Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article’s Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit http://creativecommons.org/licenses/by/4.0/.

© The Author(s) 2021